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An assessment of near surface CO_2 leakage detection techniques under Australian conditions

Andrew Feitz[a,b*], Charles Jenkins[b,c], Ulrike Schacht[b,d], Andrew McGrath[e], Henry Berko[a,b], Ivan Schroder[a,b], Ryan Noble[c,h], Tehani Kuske[a,b], Suman George[f,g], Charles Heath[f,g,h], Steve Zegelin[i], Steve Curnow[a], Hui Zhang[j], Xavier Sirault[k], Jose Jimenez-Berni[k] and Allison Hortle[c,h]

a Geoscience Australia, GPO Box 378, Canberra, ACT, 2601, Australia
b Cooperative Research Centre for Greenhouse Gas Technologies (CO2CRC), GPO Box 463, Canberra, ACT, 2601, Australia
c CSIRO Energy Transformed Flagship, GPO Box 3023, Canberra, ACT, 2601, Australia
d Australian School of Petroleum, University of Adelaide, Adelaide, SA, 5005, Australia
e Flinders University, PO Box 335 Salisbury South, SA 5106, Australia
f The University of Western Australia, 35 Stirling Highway, Crawley, WA, 6009, Australia
g National Geosequestration Laboratory, 26 Dick Perry Ave, Kensington, WA, 6151, Australia
h CSIRO Mineral Resources Flagship, 26 Dick Perry Ave, Kensington, WA, 6151, Australia
i CSIRO Marine and Atmospheric Research, GPO Box 3023, Canberra, ACT, 2601, Australia
j China Geological Survey, 1305 Qiyi Middle Road, Baoding, Hebei, China
k CSIRO Plant Industry, Cnr Clunies Ross St and Barry Dr, Canberra, ACT, 2601, Australia

Abstract

Geoscience Australia and the CO2CRC operate a greenhouse gas controlled release facility at an experimental agricultural station maintained by CSIRO Plant Industry in Canberra, Australia. The facility is designed to simulate surface emissions of CO_2 and other greenhouse gases from the soil into the atmosphere. Over 10 different near surface monitoring techniques were trialled at the Ginninderra controlled release site during 2012-2013. These included soil gas, soil CO_2 flux, soil analysis, eddy covariance, CO_2 laser, noble gas tracers, airborne hyperspectral, in-field phenotyping (thermal, hyperspectral and 3D imaging), and microbial soil genomics. Result highlights are presented. Different climatic conditions for the early 2012 release experiment (wet) and late 2013 release experiment (dry) resulted in markedly different sub-surface plume behaviour and surface expression of CO_2. The differences between the years are attributed to changes in groundwater levels and drier conditions leading to a larger vadose zone during the 2013 experiment.

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* Corresponding author. Tel.: +61-2-6249-9781; fax: +61-2-62499-920.
E-mail address: andrew.feitz@ga.gov.au

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1. Introduction

Ensuring safe and permanent storage of carbon dioxide is essential for any carbon capture and storage project. The Ginninderra project was designed to evaluate the effectiveness of different near surface monitoring techniques for detecting and quantifying leaks against a known carbon dioxide (CO_2) source. Tracking and measuring known releases of greenhouse gases in the field is the most effective approach for assessing current, and developing new monitoring techniques. The controlled release facility is an Australian first, and one of only a few similar facilities around the world [1-5]. It is designed to simulate surface emissions of greenhouse gases such as CO_2 from the ground into the atmosphere under controlled conditions of injection rate and volume. The facility has been designed and developed under a joint venture between the Cooperative Research Centre for Greenhouse Gas Technologies (CO2CRC) and Geoscience Australia, with CSIRO hosting the site at its Ginninderra Experimental Station in north Canberra (Fig. 1).

The Australian climate and soils are substantially different compared to other international controlled release facilities and near surface monitoring techniques that work in the UK, Norway or Montana (USA) may not be as equally effective in Australia. Highlights of the monitoring techniques applied under different climatic conditions are presented.

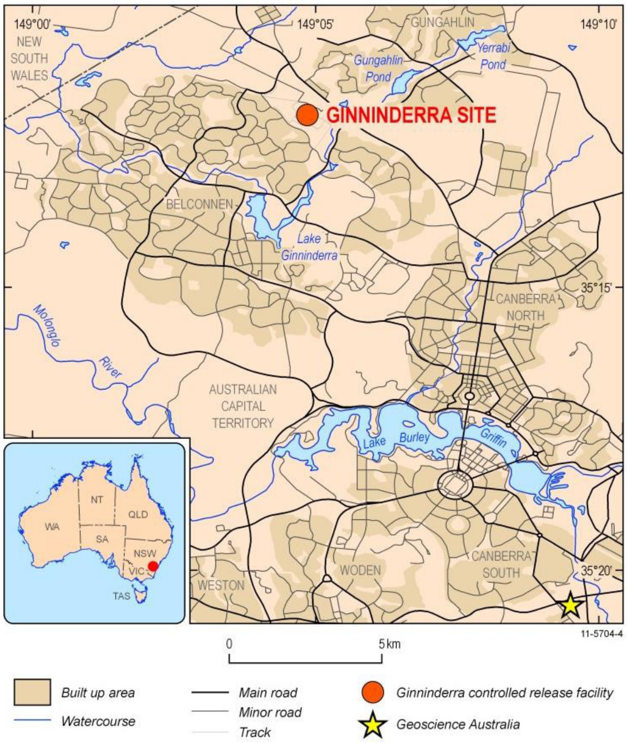

Fig. 1. Location of the Ginninderra controlled release facility in Canberra, ACT.

2. Methods

Detailed summaries of the experimental equipment and conditions are provided in Kuske et al. [6,7]. Briefly, a 2.5 tonne liquid CO_2 tank is located at the release facility and the tank was refilled every one-two weeks for the duration of the experiments. Three releases were conducted: early 2012 (Feb-May 2012; 144 kg/d for 64 days; wet season); late 2012 (Oct-Dec 2012; 218 kg/d for 56 days; dry season) and 2013 (Oct-Dec 2013, 144 kg/d for 80 days; dry season). Figure 2 shows a schematic diagram of the horizontal underground pipe installed at the Ginninderra controlled release facility. CO_2 was released from the 100 m slotted HDPE pipe, 2 m underground, from five 16 m long chambers (B–F). The well is oriented east-west. Tracers, if used, were co-injected into chamber B, which is located at the western end of the field. For the late 2012 release experiment, four different crop types were sown in 2 m wide, alternating rows perpendicular to the release well. The four crop types were field peas, canola, barley and wheat. For the 2013 release experiment, the eastern half of the field was sown with wheat and the western half with field peas, both in 2 m wide rows perpendicular to the release well.

Soil flux measurements were taken with a Westsystems CO_2 flux meter. A CO_2 analyser (LI-8100A) was used as a standalone infrared gas analyser IRGA, without a soil chamber, to sample near surface CO_2 concentrations using a tube connected to the inlet. In order to sample the gas at a constant height, the inlet tube was positioned at 5 cm from the ground level on a Trumeter survey wheel. The LI-8100A had an integrated global positioning system to map CO_2 concentration over spatial scales.

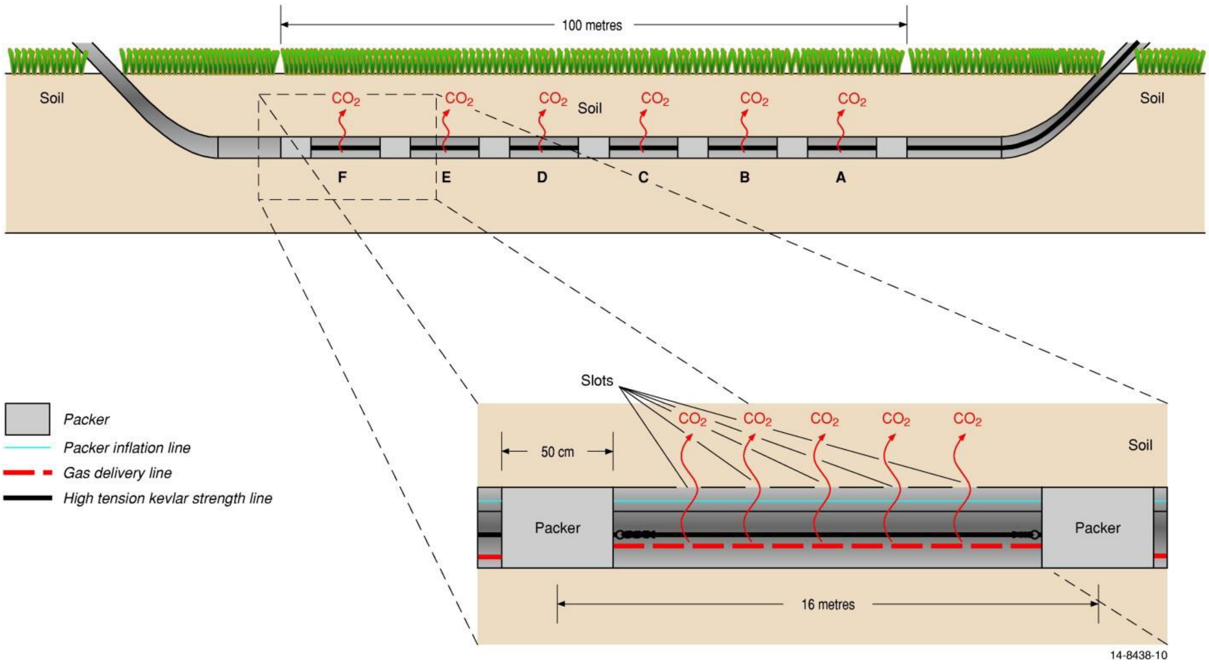

Fig. 2. Schematic diagram of the horizontal underground pipe that releases CO2 at the Ginninderra controlled release facility. Chambers are numbered A (western end - right) through F (eastern end - left).

A Post Run Tubing system (PRT) was used to access gas samples in the soil vadose zone. PRT probes were permanently installed at a depth of 1 m for the soil gas sampling, consisting of a 20 cm stainless steel screen and Teflon tubing to the surface. The wells were sealed with bentonite above the screened interval. Prior to sampling, approximately 150 mL was purged from the system via a syringe, and then a soil gas sample was pumped into a

Calibond-5 bag. Composition and tracer analysis was conducted using Gas Chromatography (GC) and Gas Chromatography – Mass Spectrometry (GC-MS).

Plant samples were taken in the field, kept on ice, and then transferred to a -80°C freezer for storage. Chlorophyll analysis and extraction was according to APHA [8] and Poorter and de Jon-Van Berkel [9].

Two eddy covariance towers [10] were used during the 2013 release experiment. Tower A comprised a Vaisala HMP50 RH & Temperature, CSI CSAT3 sonic anemometer, Li-Cor 7500 IRGA, Kipp and Zonen CNR4 radiometer, and Gill WindSonic 2D sonic anemometer. The CO_2 sensor was positioned 2.8 m above the ground surface. Tower B comprised an IRGAGSON Integrated CO_2/H_2O open-path gas analyser and 3D sonic anemometer. It also included a Gill 2D wind sonic anemometer and a temperature probe with radiation shield. The IRGAGSON was located 1.8 m above the ground surface. A Boreal Laser GasFinder2 tunable diode open path laser was used to take integrated CO_2 measurements across the plume. The laser was mounted on a programmable scanner and operated with GasviewMP software. Seven retro-reflector arrays were positioned at 1 m height at various locations across the field.

Airborne hyperspectral data in the SWIR and VNIR bands were collected using a research aircraft of Flinders University - Airborne Research Australia (ARA). A SPECIM AisaEAGLE II hyperspectral scanner (VNIR) (252 spectral bands between 400 and 1000 nm) and a SPECIM AisaHAWK (241 spectral bands between 990 and 2494 nm) were mounted in underwing pods of ARA's ECO-Dimona research aircraft VH-EOS, each one together with its own OXTS RT4003 GPS/IMU navigation and altitude system [11]. Ground-based hyperspectral measurements were taken at 2.5 m height with a Headwall Photonics 1003B-20001 hyperspectral scanner (170 spectral bands between 400 and 1000 nm). More details can be found in [12].

Three soil samples close to the greatest CO_2 flux and three others further away were sampled from 10 – 15 cm depth, before, during and after CO_2 release. All samples were measured for pH and other physicochemical properties. Soil DNA was extracted and analysed for population diversity relationships (16S RNA) using next generation sequencing and by GeoChip® analysis for nearly 4,000 genetic functional groups.

3. Results and discussion

3.1. CO_2 plant impacts

CO_2 has a visible impact on the crops and resulted in patches of yellowing or drying out of the plants above the release zone (Fig. 3 and 4). The yellowing indicates chlorosis and the inability to produce sufficient chlorophyll [13].

Fig. 3. Impact of a shallow surface CO_2 on 2013 field pea crop (left) and wheat crop (right) after 4 weeks CO_2 exposure

Canola showed an initial visible stress response to elevated CO_2 in the soil zone and the leaves turned purple. Similar plant responses at elevated CO_2 have been observed for Dandelion plants by Lakkaraju et al. [14]. The purple coloration disappeared with ongoing CO_2 exposure for the canola plants as they dried out and died. The visible extent of the impact on the plants was isolated to small sections directly above the horizontal well and typically defined a circular pattern, between 5 to 15m in diameter. This was observed in each of the crop experiments, but the location of these CO_2 affected areas changed between the experiments.

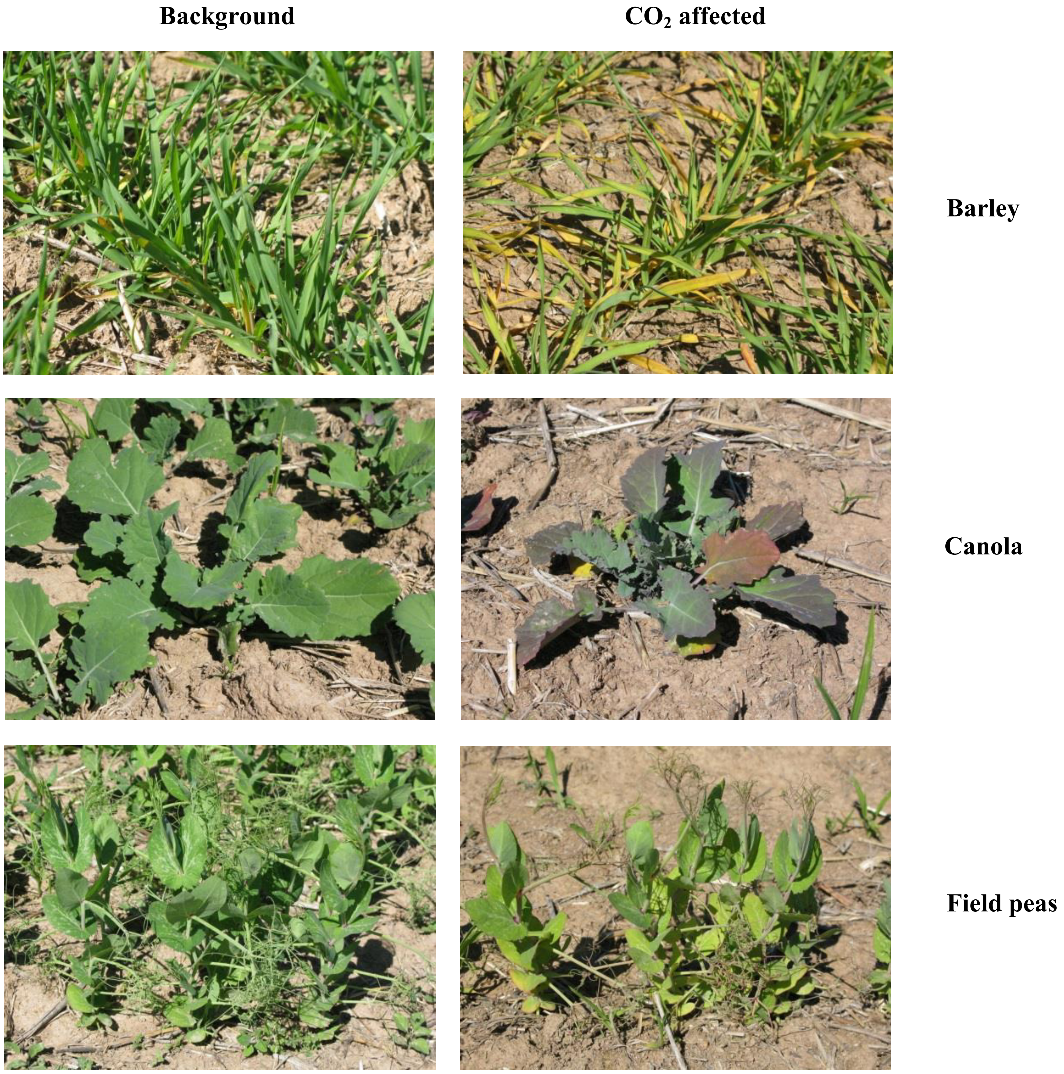

Fig. 4. Impact of a shallow sub-surface CO_2 leak on the late 2012 crops after 2 weeks of exposure to CO_2.

Compared to baseline conditions prior to CO_2 injection in the late 2013 release experiment, there is a significant reduction in the chlorophyll C content for field peas samples taken at 30 m+ from the well, some 41 and 53 % for 17 and 31 days respectively (Fig. 5). A similar, but less pronounced, reduction is observed for chlorophyll A and chlorophyll B (data not shown). This is consistent with physiology observations (e.g. proportion of green leaves;

height of plants) and a general drying of the crops towards the end of the experiment due to a combination of the crop maturity and water stress.

Within this overall reduction in chlorophyll content and general crop health, however, there is a noticeable reduction in chlorophyll content for plants exposed to high CO_2 soil fluxes. For field pea samples taken 17 days after injection, the reduction at 0 m is 50% lower compared to background samples taken at 30 m (Fig 5). For samples taken at 31 days after injection, there were insufficient viable plants at 0m to take the minimum green leaf samples required for the analysis, but between 2 – 8m from the well, there is a reduction of 30% below the 30 m background samples. Similar responses were observed in the wheat samples. There were no green leaves to analyse directly over the well but the chlorophyll C content at 2 m was 49% lower that background levels at 30 m from the well. The changes in chlorophyll C content was one of the strongest indicators of the impact of CO_2 on the crops

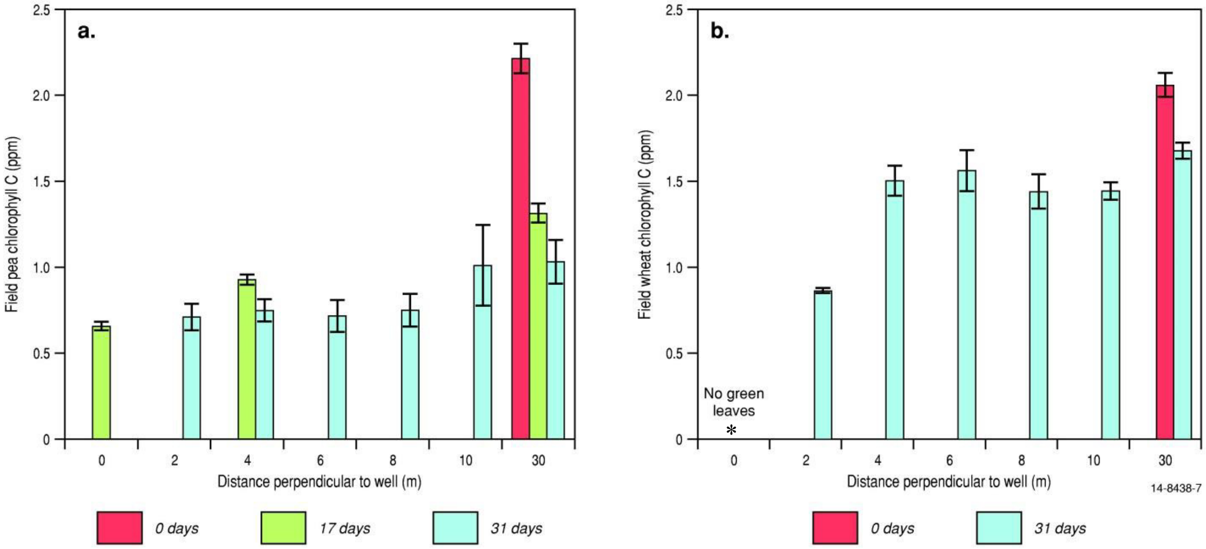

Fig. 5. Impact of a shallow surface CO_2 on chlorophyll C content for the 2013 field pea crop (left) and wheat crop (right). * No green leaves to sample after 31 days.

3.2. Airborne hyperspectral

Broad scale leak detection technologies are necessary for surveying areas beyond high risk locations and are the subject of ongoing research. Airborne hyperspectral and thermal scanning measurements were taken over CO_2-impacted, mature wheat and field pea crops in 2013. Dry conditions and the late maturity state of the crops strongly influenced the effectiveness of the remote sensing techniques for CO_2 leak detection. The generally dry conditions of the crops meant it was difficult to differentiate between water/heat stressed plants and plants additionally affected by CO_2. However, preliminary analysis indicates that some vegetation indices (Fig. 6) show some promise. Low responses values for the chlorophyll normalized difference index (Chl NDI) are coincident with locations of elevated CO_2 surface soil flux in the field and these have a more diffuse shape compared to the strong dark lines for roads, buildings, spectral ground targets and other structures nearby. Fields containing pasture above, to the left and right of the cropped fields do not have these red spots. Unfortunately, a similar low response is observed in the adjacent field below, which contains a dead field pea crop. This emphasizes the challenges of reducing false positives; however, through a combination of suitable indices, possibly shape classification (i.e. looking for circular features), and aerial photography, greater selectivity may be possible. Further advances in hyperspectral measurements and image processing could provide to the capacity for CO_2 leak detection of wide areas at relatively low cost. Patterns of plant stress due to natural causes are likely to repeat year by year, being linked for example to soil fertility and drainage, whereas damage caused by CO_2 will not necessarily follow these patterns in space and time.

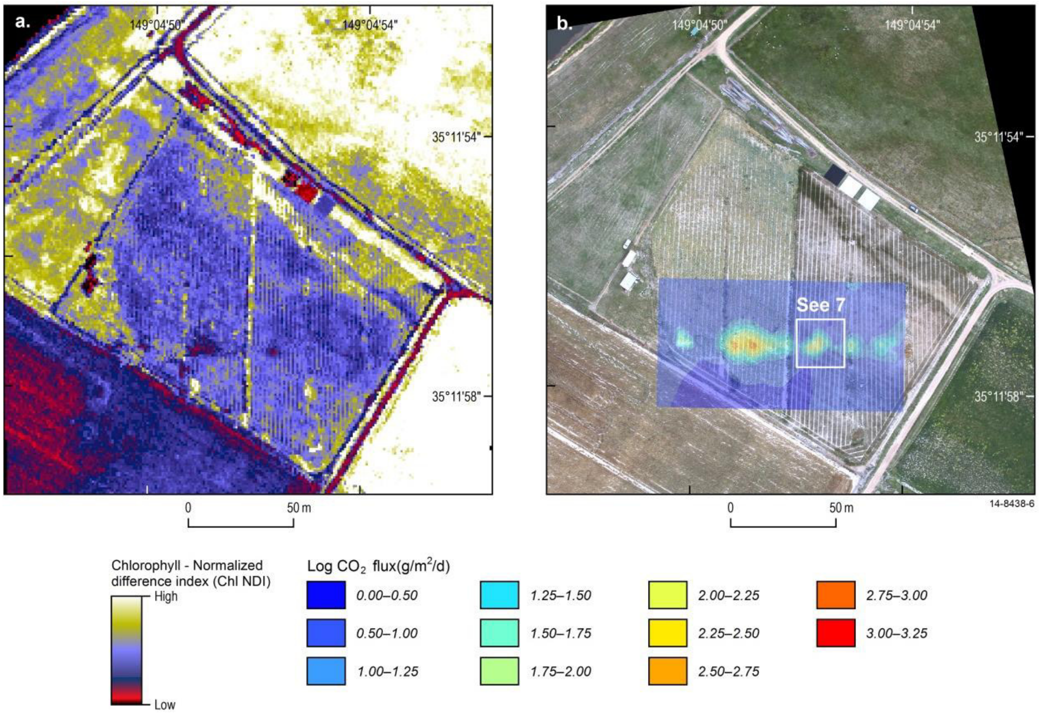

Fig. 6. Comparison between (a) Chlorophyll normalised difference index (Chl NDI); and (b) CO_2 soil flux map overlaid on aerial photo taken on the 3/12/13.

3.3. Phenomobile hyperspectral

A comparison between high resolution ground-based hyperspectral measurements taken during 2012 with the Phenomobile [7] and hyperspectral measurements obtained using an aircraft is presented in Figure 7. The Phenomobile acquired hyperspectral imagery approximately 2.5 m above each row of crops between the 60 m and 90 m soil gas transects (Fig. 10). While the high resolution data coverage was less than that obtained from the aircraft, much greater discrimination is apparent within the Phenomobile survey extent, particularly between different plant species (Fig. 7). The canola in particular shows a strong Chl NDI response and the location of the leak is clearly visible the central row (i.e. central black area). The wheat and barley crops had a much lower Chl NDI response due to the lower ground cover compared to the canola and field peas (Fig. 8). These results provide a preview into the degree of resolution that may be possible using remote systems in the future.

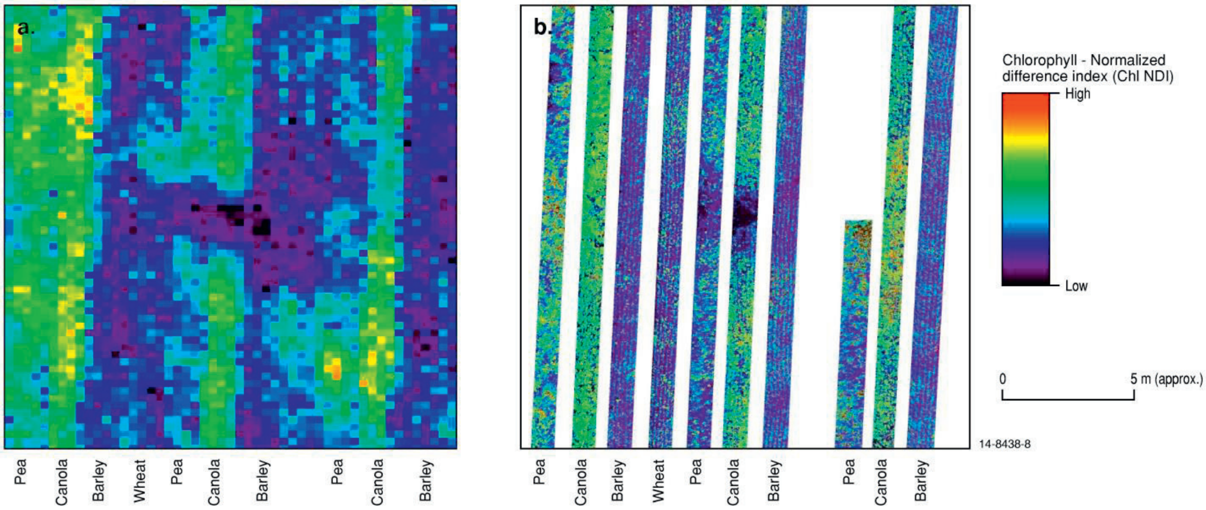

Fig. 7. Comparison between hyperspectral images taken at the same location near the high surface CO_2 flux zone between the 60 and 90 m soil gas well transects using (a) airborne hyperspectral (b) the Phenomobile during 2012. Chlorophyll Normalised Difference Index (Chl NDI) values are displayed. Both images cover the same extent (see white box in Fig 6b for the location within the field). Please note that this figure shows hyperspectral data from 2012 whereas figure 6 shows hyperspectral data from 2013. The hyperspectral measurements taken using the phenomobile have much greater resolution and individual plants can be visualised.

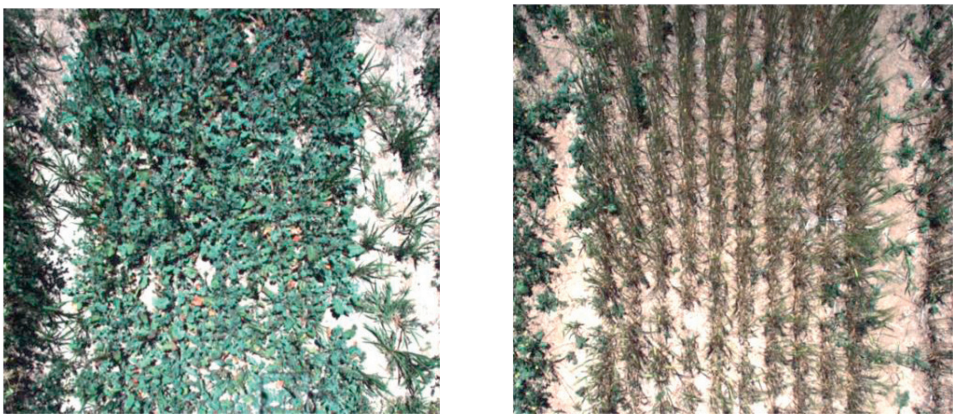

Fig. 8. Example photos illustrating the much greater ground cover for canola (left) compared to barley (right). Photos were taken 2.5m above each row using the Phenomobile.

3.4. Surface CO_2 concentration map

Geo-referenced near-surface CO_2 concentration measurements using a LICOR CO_2 analyser and Trumeter survey wheel were used to locate CO_2 surface leaks (Fig. 9). Manual traverses across the field, up and down the crop rows (2.2 m apart), provided a rapid assessment tool for identifying aberrations in CO_2 concentration relative to background level at high resolution. The measurement of CO_2 concentrations were determined by sampling at a constant height above the soil surface (5 cm) and proved to be successful provided the wind speed was low (measurements were taken early in the morning). This technique detected CO_2 leaks earlier than soil flux measurements and a detailed survey across the 1.5 Ha field could be completed within 2 hours.

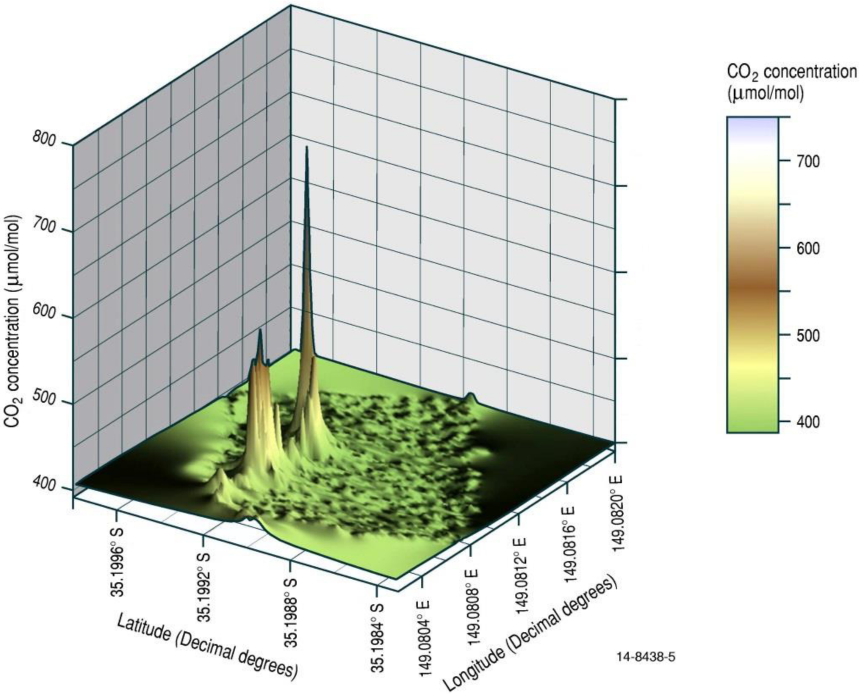

Fig. 9. Surface CO_2 concentration map during the 2013 release

3.5. CO_2 soil flux

The most obvious difference between the early 2012 and 2013 release experiments was that CO_2 leakage expressed at different locations along the well for the different experiments. CO_2 was released at the same rate for both experiments. As observed in other controlled release experiments internationally [15,16], the surface expression of CO_2 during these experiments, as measured using a portable soil flux meter, was restricted to localized areas. The surface expression at Ginninderra was typically less "patchy" than other controlled release sites that have greater sand and gravel sediment content in their soils [15-17]. For the 2012 (wet) release experiment, the leakage was limited to an intense primary leak zone (approximately 16m x 30m) (Fig. 10a). In contrast, the leak from the 2013 (dry) release experiment was spread in three smaller spots over a longer length of the release well and did not attain the very high flux intensities observed in the previous year (Fig. 10b). These results underscore the need for broad-scale detection technologies over kilometer scales to first detect small scale features before they can be mapped and quantified using soil flux techniques [17].

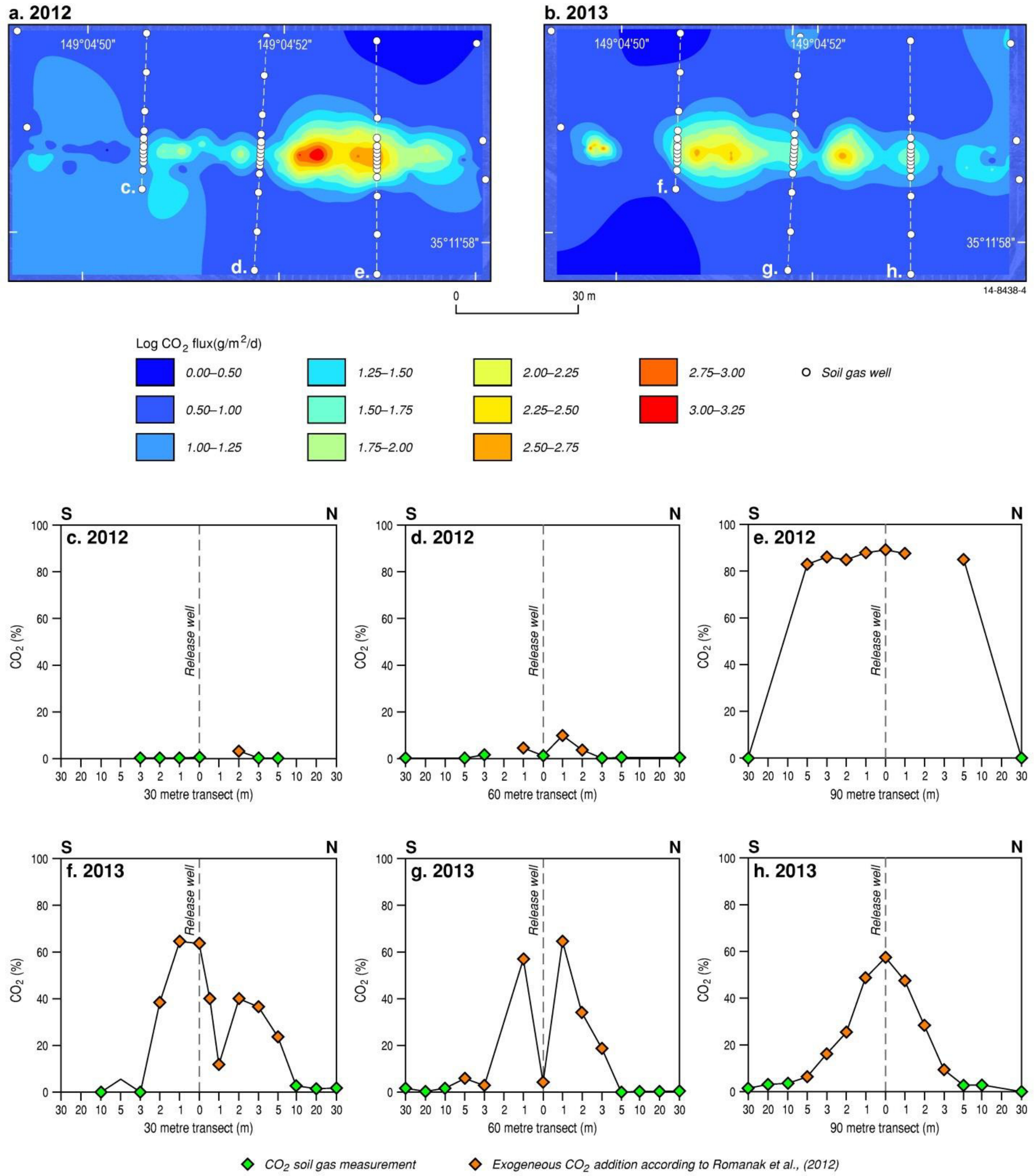

Fig. 10. A comparison between surface CO_2 soil flux and CO_2 soil gas (1 m deep) for the early 2012 release experiment (wet) versus the late 2013 release experiment (dry). Release rate for both experiments was 144 kg/d. Soil gas transects across the well occur at 30, 60 and 90 m from the western end of the horizontal well. Elevated CO_2 concentration in the soil gas wells that are consistent with exogenous CO_2 [18] are highlighted in orange.

3.6. Soil gas concentration

An array of 1 m deep soil gas wells provided insight into the migration pathways of CO_2 in the sub-surface, showing a much broader dispersion of CO_2 in the sub-surface compared to the surface CO_2 expression. For the early 2012 (wet) release, CO_2 surface expression was restricted to the eastern part of the horizontal well (Fig. 10c-e). Very high CO_2 concentrations are observed in the soil gas wells out to 5 m (>80%, Fig. 10e). In comparison, CO_2 concentrations in the soil gas wells under dry conditions was much more evenly distributed along the horizontal well and rarely exceeded 60% (Fig. 10f-h). Krypton tracers confirmed that the spread of the introduced gases in the sub-surface was much greater than the surface expression, with different behaviour observed between the 2012 and 2013 experiments. Krypton was observed up to 30 m perpendicular to the well at 1 m depth during 2012 but only 5-10 m during the late 2013 release experiment. The differences between the years are attributed to changes in groundwater levels, drier conditions, and a larger vadose zone during the 2013 experiment.

Fixed soil gases such as O_2 and N_2 were also investigated, as a newly developed approach by Romanak et al. [18] examines several relationships among major fixed gases (CO_2, O_2, N_2) to determine the soil gas origin. Based on this approach samples ranged from being not or only little affected to strongly affected by the release experiment. They are marked green and orange respectively in Figure 10c-f. Those wells with elevated CO_2 were all identified as exogenous CO_2 additions using the soil gas ratios.

3.7. Eddy covariance

Eddy covariance (EC) towers were deployed at the site for different experiments with the objective to detect and quantify CO_2 emissions. During the 2013 experiment, two EC towers were installed at the site. Tower A was approximately 56m SEE from the centre of the main plume whereas tower B was 33m SE of the main plume (Fig. 11). The closer Tower B was operational for only 2 weeks during the release. CO_2 leaks were detected above the background and the direction of the leak confirmed independently for both towers (Fig. 12). However, analysis showed that current methods of EC are not appropriate for quantifying the CO_2 leak, as much of the CO_2 flux is lost through advection and diffusion below the measurement height. This is because the footprint of the leak is much smaller than the footprint that the EC tower can measure, resulting in a highly heterogeneous system.

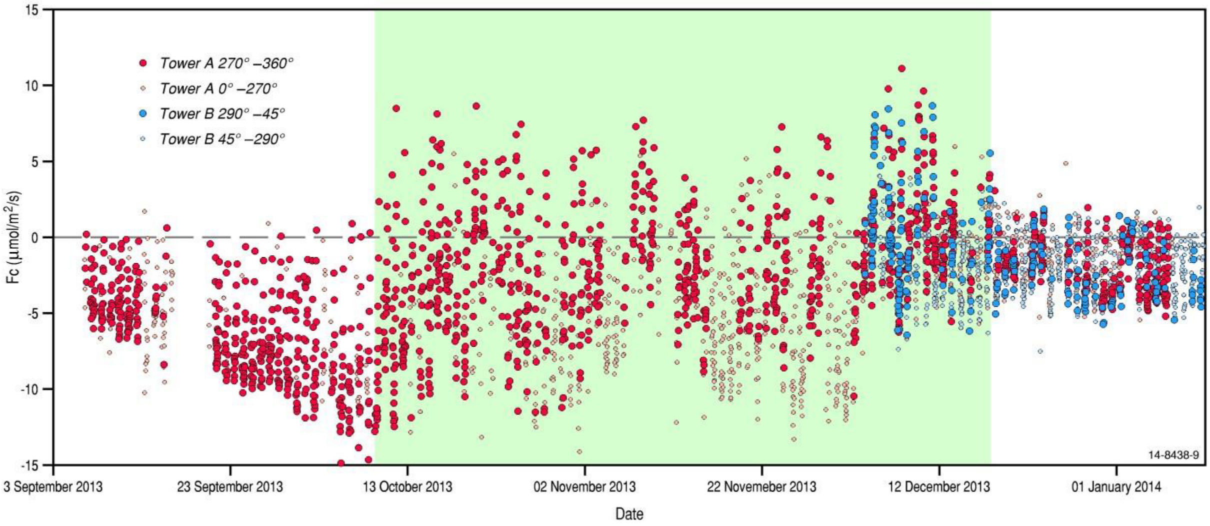

Fig. 11. Eddy covariance measurements of CO_2 flux using two towers during 2013. Data is filtered for daytime periods. Negative fluxes indicate strong uptake of CO_2 during the day due to photosynthesis. The green region indicates the CO_2 release period.

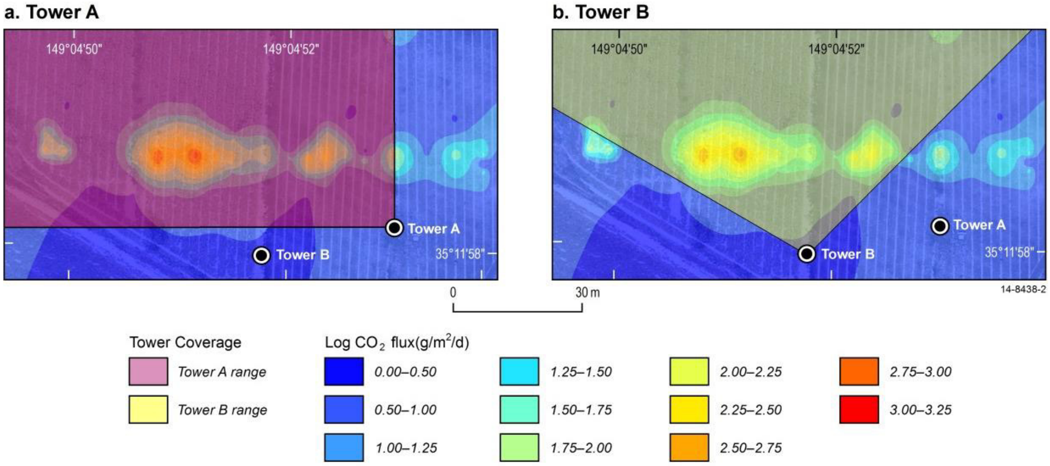

Fig. 12. Layout of eddy covariance towers and their assigned direction range for detection (Tower A = 270° to 360°; Tower B: 290° to 45°).

3.8. Open path laser CO_2 measurements

A Boreal open path laser was used to scan across the plume in the field on 17 December 2013, using seven reflectors all at the 1 m height The integrated CO_2 path responses are consistent with the surface expression of the leaks and mapped CO_2 soil flux. Reflectors 4 and 7 show the highest path-averaged CO_2 perturbation above background (18 and 22 ppm, respectively). These are located downwind of the major leak site and smaller leak to the east (Fig. 13). R2 crosses the western edge of the primary leak but no significant signal is observed, suggesting the plume originating from the ground is below the line of the laser. A 7 ppm perturbation is observed along path R1, downwind of the most western leak. These preliminary results indicate that it is possible to detect a perturbation above the background signal. The measurements could be potentially combined with plume dispersion models and Bayesian statistics to estimate the CO_2 leakage rate [19,20,21].

3.9. Electromagnetics

Measurements of soil conductivity were made with a Geonics EM31, which measures soil conductivity by an inductive method. The effective depth of investigation either 3 or 6 m, and the survey data were logged with real-time GPS co-ordinates at 1 m height. Surveys were made before and during CO_2 release for both the late 2012 and the 2013 releases. Despite the differing soil saturation on these two occasions, the patterns of soil conductivity were very similar. There were no changes in conductivity above a noise level of about 5%, except for a leaking water pipe which was convincingly detected (this leak also showed up in the thermal infrared hyperspectral imagery). The pattern of conductivity appears to be caused by fluvial gravel deposits, linked to a pre-agriculture creek bed. However there are no apparent small-scale features which might be associated with higher permeability and the development of the patchy flux pattern (Fig. 10). The dissolution of CO_2 in groundwater would be expected to be undetectable at the level of repeatability of our measurements (2 – 3 mS/m).

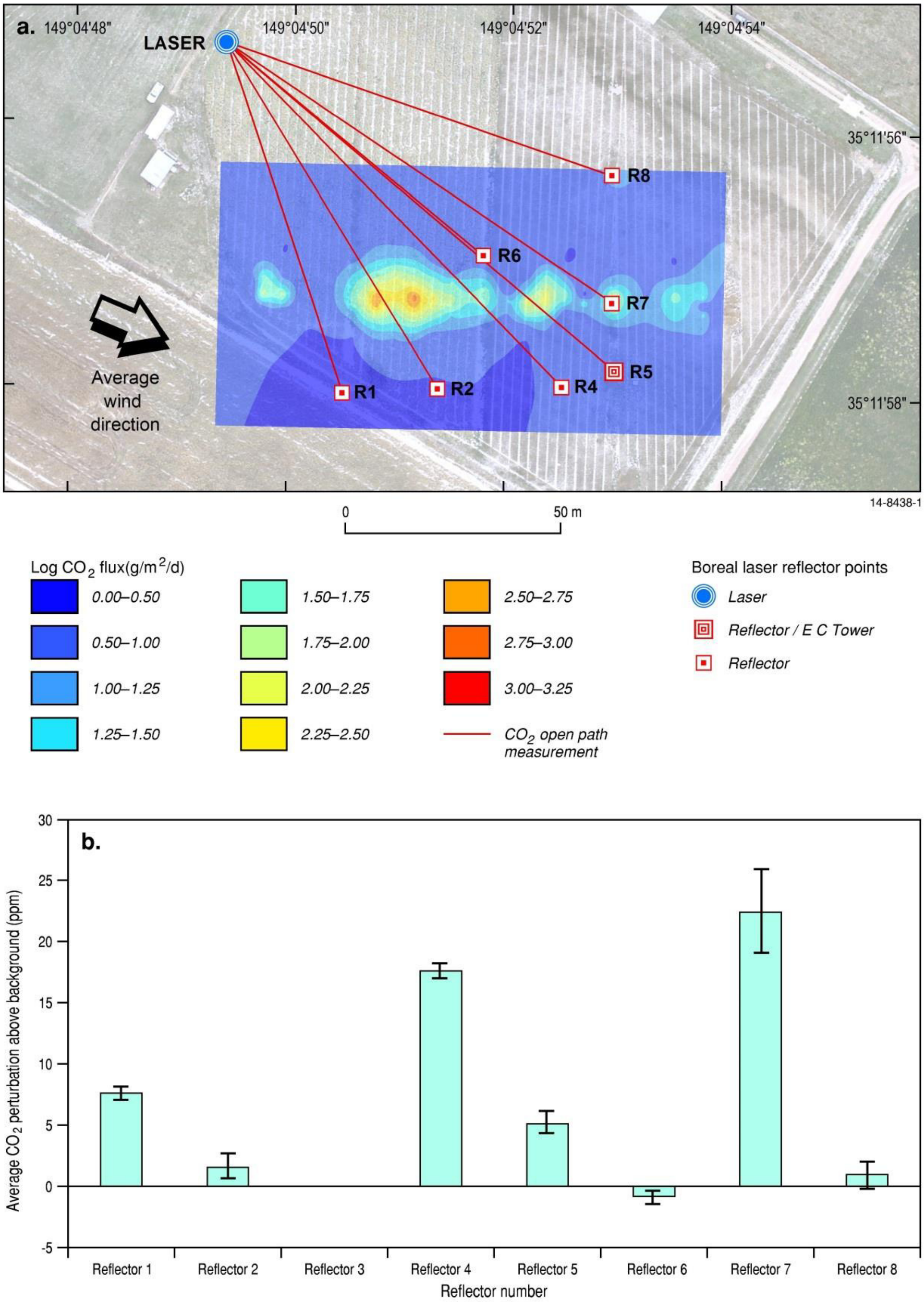

Fig. 13. The figure shows (a) the open path CO_2 laser measurements overlaid on an aerial photo and soil flux map, and the location of the reflectors; and (b) the average perturbation above background for each of the paths.

3.10. Microbial surveys

The results show a clear shift in the microbial community related to a switch from aerobic to anaerobic respiration near the CO_2 leak during the early 2012 release experiment. This shift can be isolated from even stronger seasonal influences by statistical analyses. *Nitrospira* and *Firmicutes* phyla increase in numbers after the CO_2 increase. These species are anaerobic, more acid/metal tolerant species which is confirmed with the increase in functional genes related to metal resistance and bioleaching in the GeoChip® results. The pH was not observed to change significantly and is buffered by the soil and the microorganisms.

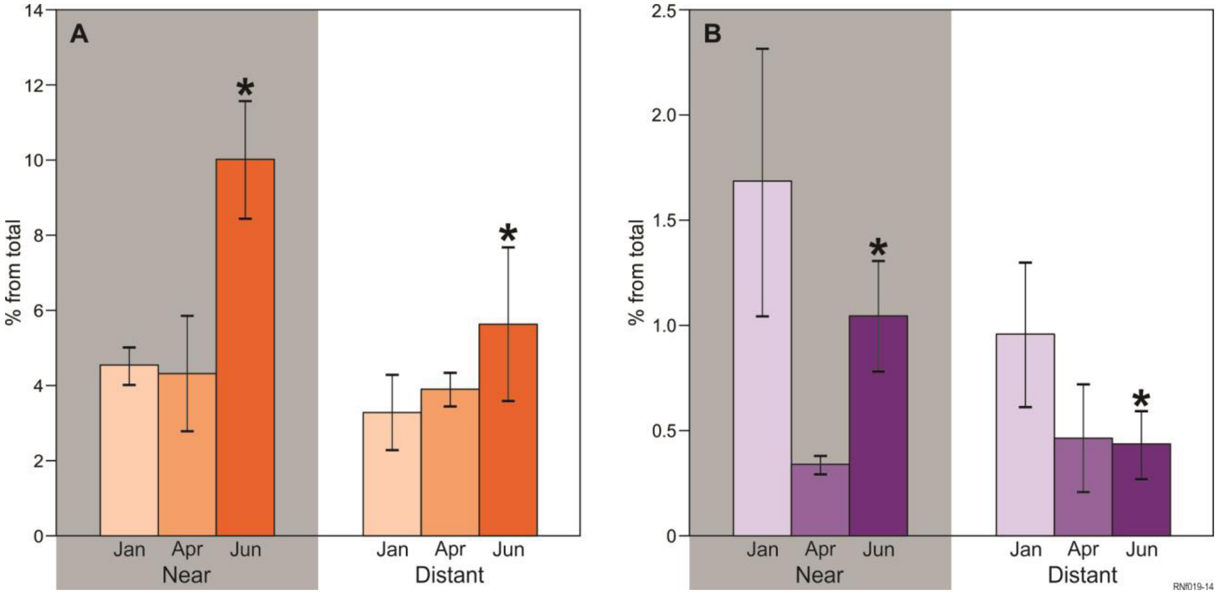

Fig. 14. Abundance (Percent from total number of reads in the sample) of *Firmicutes* (A) and *Nitrospira* (B) in soil samples near (shaded) the leak and at a distance. The abundance is given as a mean of 3 replicates. Error bars represent the standard deviation of the mean. * indicates significant difference ($p<0.05$).

4. Conclusions

- Surface expression of CO_2 is not uniformly distributed, even under highly controlled conditions, rather it is localised and patchy, expressing as small (1-16m in diameter) high intensity flux “hot spots”. This is consistent with observations conducted internationally at natural CO_2 seepage sites and other controlled release studies.

- The location of the surface expression is also dependant on climatic conditions, most likely due to the influence of groundwater levels and the extent of the vadose zone.

- The lateral extent of the subsurface soil gas footprint is greater than the CO_2 soil flux surface expression, particularly under wet conditions.

- CO_2 impacts on plants can be clearly observed at ground level. There is some evidence that similar CO_2 impacts can be differentiated against other plant stresses (heat/lack of rain) using airborne hyperspectral measurements but the application is currently limited by the high level of false positives.

- EC was able to detect CO_2 leaks above the background CO_2 flux, however existing methods are currently not suitable for accurately quantifying leaks due to the small, localised nature of leaks relative to the area being measured.

- It is possible to detect and possibly quantify leaks using an open path CO_2 laser scanning over a large area with several reflectors.

- Microbial surveys show a shift in the microbial community related to a switch from aerobic to anaerobic respiration near the CO_2 leak.

- The primary monitoring challenge is to detect small-scale leak features over large areas and to reduce the number of false positives.

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