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Atmospheric monitoring of the CO₂CRC Otway Project and lessons for large scale CO₂ storage projects

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Abstract

Monitoring is essential for the approval and control of geological storage of carbon dioxide and to judge the effectiveness of the technology in mitigating CO₂ emissions and climate change. We present a strategy for monitoring the atmosphere in the vicinity of a geological storage project that is designed to detect and quantify potential emissions. The strategy includes measurements of CO₂, CO₂ fluxes and tracers, combined with model simulations of atmospheric dispersion and ecosystem CO₂ fluxes. We applied an atmospheric monitoring program to the CO₂CRC Otway Project where large amounts of CO₂ have been stored in a deep depleted natural gas reservoir. The sensitivity of the monitoring is tested by detecting emissions from surface activities at the Otway facility, including a scheduled release of injected gas.

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1. Introduction

Evidence that geologically stored CO₂ doesn't escape to the atmosphere is increasingly required by project operators, by regulators of health, safety and environment, for national carbon accounting obligations, by carbon markets and by the public. Public support for carbon capture and storage (CCS) is one of the main requirements for investment into this technology and the key public concern regards leakage.

The risk of leakage is extremely small for well characterised sites and carefully managed and monitored injection. Leakage is defined here as the emission of gases to the atmosphere, including the injected CO₂, gases that might accompany the injected CO₂ and gases such as methane that might be displaced from the storage reservoir.

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Health and safety guidelines usually require that concentration thresholds of some gases in the air are not breached, for example, about 1% mole fraction for CO₂. Such situations would result only from extremely high leakage rates which can be readily monitored. For carbon storage to provide effective mitigation of climate change, however, leakage rates will need to be extremely small. Verifying that such leaks do not occur poses a challenge for conventional atmospheric monitoring techniques, especially for CO₂ because the local concentration increases due to small leak rates would typically be much less than the variations caused by ecological fluxes and anthropogenic sources.

While a thorough storage monitoring program would involve verification of CO₂ containment in the sub-surface target formation, atmospheric monitoring can be non invasive, is relatively inexpensive and can be feasibly carried out in a continuous, operational manner over many years or decades. Atmospheric monitoring also has the potential to detect, locate and quantify leakage (Lewicki et al., 2005 [1]; Leuning et al., 2008 [2]; Loh et al., 2009 [3]).

Here we present an atmospheric monitoring strategy that has been devised to detect and quantify potential leakage of geologically stored CO₂ and show how it has been applied to the CO2CRC Otway Project in Australia (<http://www.co2crc.com.au/otway/>). The Otway Project has injected about 65,000 tonnes of fluid (approximate molar composition 80% CO₂ and 20% CH₄ and minor amount of tracers such as SF₆) into a depleted natural gas reservoir between March 2008 and August 2009 and has a sufficiently large scale to approximate the main aspects of a commercial CCS project. Atmospheric monitoring is part of a comprehensive monitoring program that includes seismic surveys, geochemical measurements of the reservoir (via a deep well), groundwater and soils.

2. Climatically tolerable leak

The ultimate goal of atmospheric monitoring, from a climatic perspective, is to assess whether CCS can withhold CO₂ from the atmosphere sufficiently well to mitigate climate change. Several modelling studies have shown that sustained climate benefit from CCS requires that leakage is kept below a maximum rate of about 0.1% to 0.01% of the stored CO₂ per year (Haughan and Joos, 2004 [4]; Enting et al., 2008 [5]; Shaffer, 2010 [6]). The range derives partly from a number of considerations and assumptions in the modelling, such as the leakage being globally-averaged and beginning from the start of injection, and the energy penalty (and extra CO₂ production) associated with capture and storage. Geological considerations suggest that such low leakage should be possible. The IPCC, for example, arrives at similar estimates, that 99% retention over 100 years is very likely for geological storage of CO₂ under optimum conditions (Metz et al., 2005 [7]).

Applying the relative leak of 0.01% per year to a nominal 10 Mt CO₂ commercial storage gives 1000 t CO₂ per year which we define as the target leak for the development and assessment of our monitoring.

3. Simulations and strategy

We based our measurement strategy (Etheridge et al., 2005 [8]; Leuning et al., 2008 [2]) on the simulated changes in atmospheric composition resulting from hypothetical leakage from the Otway Project (Figure 1). We used the CSIRO atmospheric dispersion model TAPM (Hurley et al., 2005 [9]; Luhar et al., 2009 [10]) to calculate the concentrations of CO₂ and other gases when hypothetical emissions were released from a point source and from a diffuse source (1x1 km) centered on the injection well (CRC-1). The target leak of 1000 t CO₂ yr⁻¹ was used in simulations even though it would be unrealistically high for the much smaller amount stored at Otway. Synoptic meteorological analyses were used as boundary conditions and simulations were run for January and August 2004. The Otway site is relatively flat pastoral land about 4 km inland of the southwest Victorian coast and has no major towns or busy roads nearby. Potentially significant anthropogenic sources of CO₂ and other trace gases are agriculture (including cattle), occasional burning off and a commercial CO₂ extraction plant 1.1 km east of the atmospheric facility. However, the overwhelming source of variability in CO₂ concentrations is the biospheric CO₂ flux from the surrounding pasture ecosystem.

The location of our monitoring station was chosen to balance the competing requirements of signal size, which is larger near a leakage source, with the probability of intercepting a plume when the location of the source(s) is unknown. The composition measurements from a point inlet are preferred to open path measurements because the signal of a plume measurement can be reduced by the line averaging of the open path system (Loh et al., 2009 [3]). Ideally, a dense network of continuous, precise and intercalibrated analysers for several compounds would be

installed across the site (Loh et al., 2009 [3]; Jenkins et al., 2010 [11]). Alternatively, frequent mobile surveys across the land surface might identify leakage “hot spots” (eg. Hirst et al., 2004 [12]; Krevor et al., 2010 [13]). However, such approaches might be costly and impractical, requiring land access for many years across sites that might be many square kilometres in size. The Otway atmospheric measurement station is therefore situated about 700 m from the injection well and downwind of the storage site during winds from the southwest-southeast sector, when the air is unaffected by regional gas sources. The layout of the atmospheric measurements is shown in Figure 1. With this configuration, the composition of air at the baseline air pollution station at Cape Grim (<http://www.bom.gov.au/inside/cgbaps/>) in northwest Tasmania approximates an upwind boundary condition. Simulated perturbations above the background of up to 3 ppm CO₂ were found for the target leak (Etheridge et al., 2005 [8]; Leuning et al., 2008 [2]). For gas containing 1 micromolar SF₆ added tracer, simulated leakage perturbations (up to 3 parts per trillion, ppt) would be much easier to detect than for CO₂ alone because of the low and relatively steady concentrations of SF₆ in the background atmosphere.

Although the simulations were configured for Otway, they could be broadly representative of other storage sites in flat grassed areas under comparable wind conditions.

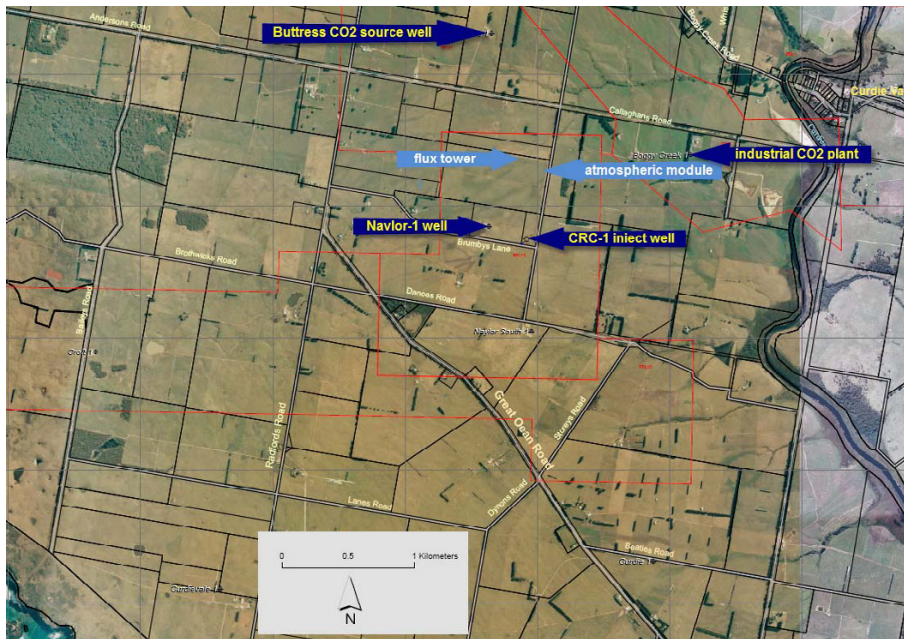


Figure 1. Map of the Otway project site (adapted from a CO2CRC image) showing the location of wells (Buttres 1 source well, CRC-1 injection well, Naylor-1 observation well) and other potential gas sources (town, roads, CO₂ plant) and the atmospheric measurement set up. The location of the atmospheric module is 38° 31.52' S, 142° 48.81' E and is about 3.5 km northeast of the Victorian coast (shown bottom left).

4. Measurements

Continuous measurements of CO₂ concentration began in January 2007, using a CSIRO LoFlo analyser (Francey and Steele, 2003a [14]). The measurements (Figure 2) show large variations compared to the “baseline” CO₂ concentrations measured at Cape Grim. These are mainly due to local ecosystem CO₂ fluxes. High concentrations at night time result from a combination of respiration CO₂ emissions and stable atmospheric conditions (weak dispersion). Low CO₂ concentrations (often lower than for the baseline atmosphere) correspond with photosynthetic uptake and stronger mixing. During strong winds, especially from the south (off the ocean) the concentration of CO₂ (and other trace gases) is very similar to that at Cape Grim.

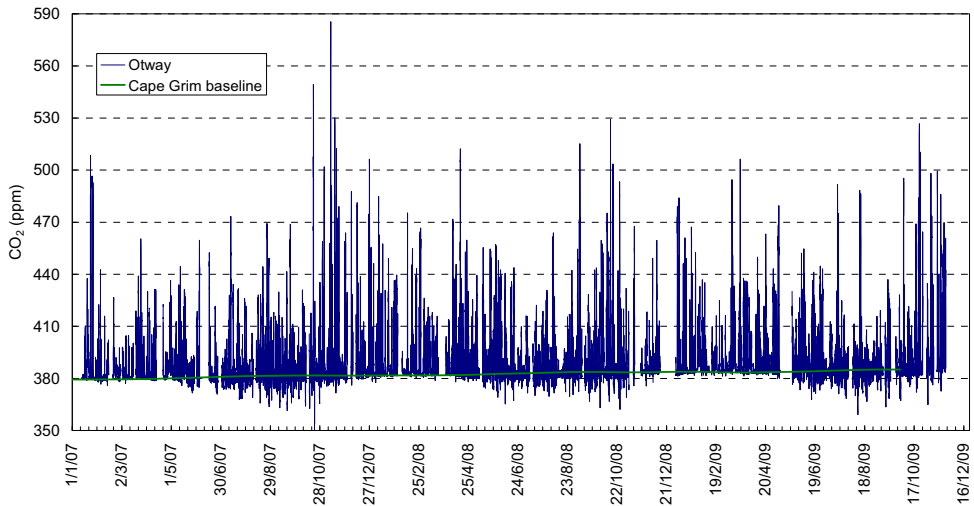


Figure 2. CO₂ concentration record (hour mean mole concentrations in dry air) from the CSIRO LoFlo non-dispersive infrared (NDIR) analyser at the Otway Project atmospheric station (from an inlet at 10 m height) and for the Cape Grim Baseline Air Pollution Station (selected for baseline wind conditions). Measurements are calibrated to the World Meteorological Organisation X2007 mole fraction scale.

Land-air CO₂ fluxes are measured above the subsurface reservoir using the eddy-correlation (EC) flux tower technique (Leuning et al., 2008 [2]). Flux and meteorological data from the flux tower are also used in the model simulations of the ecosystem CO₂ fluxes and atmospheric dispersion at the site. To complement the interpretation of the flux tower data, soil CO₂ fluxes were measured during several campaigns before and after CO₂ injection at randomly selected points across the area. The EC CO₂ fluxes are shown in Figure 3.

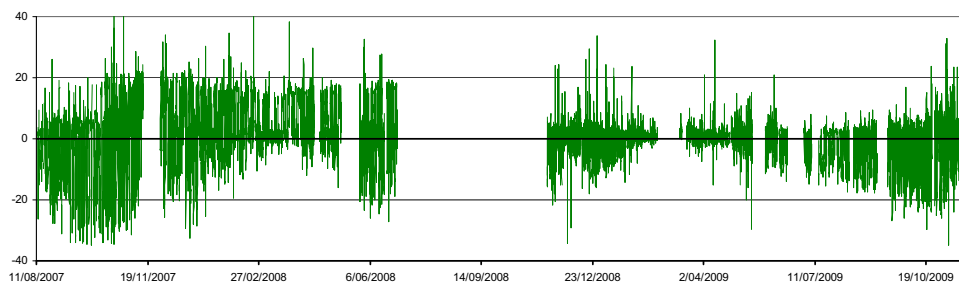


Figure 3. CO₂ fluxes in $\mu\text{mol CO}_2 \text{ m}^{-2} \text{ s}^{-1}$ from the EC flux tower (hour means at a height of 4 m). Negative values indicate CO₂ uptake by the land surface.

A seasonal cycle is apparent in both CO₂ concentrations and fluxes. Maximum ecosystem activity (CO₂ uptake by photosynthesis and emission from respiration) occurs during winter and spring (July to November) while there is a reduced range (and almost no photosynthesis) during the dry conditions of late summer and autumn (February to May). The relatively low background variability during the dry period would improve the potential of detecting CO₂ signals from sources such as leakage. CO₂ injection began at the end of March 2008.

Ideally, modelled CO₂ concentrations should closely match the measured record when ecological fluxes, either measured by the flux tower or modelled by the land-surface scheme “CABLE” (Community Atmosphere Biosphere Land Exchange; Kowalczyk et al., 2006 [15]) and other known local and regional sources are dispersed in TAPM. Underestimates would then identify unaccounted sources, such as leakage. However, uncertainties in the modelling, especially at night, are presently too high to predict the CO₂ concentrations with sufficient accuracy to reveal anomalies that might indicate a leak at the small target rate described earlier (Luhar et al., 2009 [10]).

This limitation is largely due to the relatively high background CO₂ concentration and variability, particularly in an ecosystem such as Otway. Enhancements would be difficult to detect in CO₂ concentration alone. In a separate study, Loh et al. (2009 [3]) measured and modelled the controlled release of gases in a field experiment and found that increases above background concentrations of 1% or more were required to accurately estimate the emission rate of a gas source using concentration measurements. Detection and quantification of leakage can therefore be improved by monitoring gases that accompany the stored fluid as natural or introduced tracers and that have relatively low and stable background concentrations in the atmosphere. There are a number of potential tracers at Otway and these are a key part of the Otway atmospheric monitoring program. Methane (CH₄) comprises about 20% of the injected gas. It is also present as residual gas in the storage reservoir and would make a useful tracer of gas that is displaced by the injected fluid. Sulfur hexafluoride (SF₆) is one of several introduced tracers. A useful natural tracer at Otway is ¹³CO₂. The injected gas is typical of magmatic CO₂, having an isotopic ratio ($\delta^{13}\text{CO}_2$) of -6.8 per mil (‰). This is substantially different from the local ecosystem $\delta^{13}\text{CO}_2$ that dominates the atmospheric CO₂ variations through respiration and photosynthesis. Measurements of CO₂ and $\delta^{13}\text{CO}_2$ during variations over several months give a mean value of -28 +/-1 ‰, typical of a C3-photosynthesis dominated ecosystem. Monitoring of $\delta^{13}\text{CO}_2$, CH₄, SF₆ and other trace gases is therefore a key part of the Otway atmospheric program.

Continuous measurements are highly advantageous in detecting leakage signals (Etheridge et al., 2005 [8], Loh et al., 2009 [3]). Recent advances in spectroscopic measurement techniques (wavelength scanned cavity ring down spectroscopy, WS-CRDS) are providing instruments capable of precise, continuous measurements of several gases at atmospheric levels and suitable for field deployment. Measurements of CO₂, CH₄ and $\delta^{13}\text{CO}_2$ have recently begun at the Otway atmospheric station using two Picarro WS-CRDS instruments (Picarro Inc.). In addition, measurements of CO₂, CO₂ stable isotopes, CH₄, CO, H₂, N₂O and halogenated gases such as SF₆ have been made since the start of the project via flask air samples and subsequent laboratory analysis (Francey et al., 2003b [16]). Samples are taken at approximately monthly intervals or more intensively during periods of interest (such as scheduled surface emissions).

5. Detection of emissions

We found no evidence for changes in concentrations of CO₂ or tracers, isotopes or CO₂ fluxes that together would indicate leakage from the subsurface to the atmosphere at the Otway project. Monitoring of the subsurface (Jenkins et al., in preparation [17]) confirms that the injected fluid behaved as predicted and stayed within the target reservoir. Near surface monitoring also found no evidence of the injected fluid in soil gases or aquifers. In the absence of leakage, we focus on periods when emissions from surface activities were likely to have affected atmospheric composition. We use measurements and modelling during three events to demonstrate the atmospheric monitoring strategy and to calibrate its sensitivity. The modelling results are shown only for one event.

In that event, emissions were caused by the scheduled venting of gas from the observation well (Naylor-1) during a geochemical sampling and measurement campaign on December 8, 2009. The gas molar composition included 63% CH₄, 29% CO₂ and 3.5 parts per million SF₆. Venting of about 20 kg hr⁻¹ occurred for 90 minutes in the morning, when the wind was from the southwest, carrying the vented gas toward the atmospheric station. The CO₂ emission rate (about 85 t CO₂ yr⁻¹) was about one-tenth the target leak rate. In contrast, the CH₄ emission rate (about 67 t CH₄ yr⁻¹) would be comparable to the target leak rate with 20% mole fraction CH₄ in the fluid, as injected at

Otway. The SF₆ emission rate was comparable to the target leak rate containing SF₆ tracer at about 1 ppm mole fraction.

Figure 4 shows the atmospheric measurements during the day when venting occurred. CO₂ concentrations measured at the atmospheric station showed the typical diurnal pattern: high during the night and low during the day. The flux tower measurements show that this was caused by CO₂ emissions at night and uptake during the day, consistent with respiration and photosynthesis respectively. Daytime concentrations did not exceed the baseline concentration at Cape Grim during the venting period and were as much as 4 ppm below. Elevated CO₂ concentrations from the well venting were expected to be small and were masked by the large ecosystem uptake and variable dispersion. Concentrations of CH₄ and SF₆, however, were significantly enhanced during venting, compared to their relatively low and stable backgrounds.

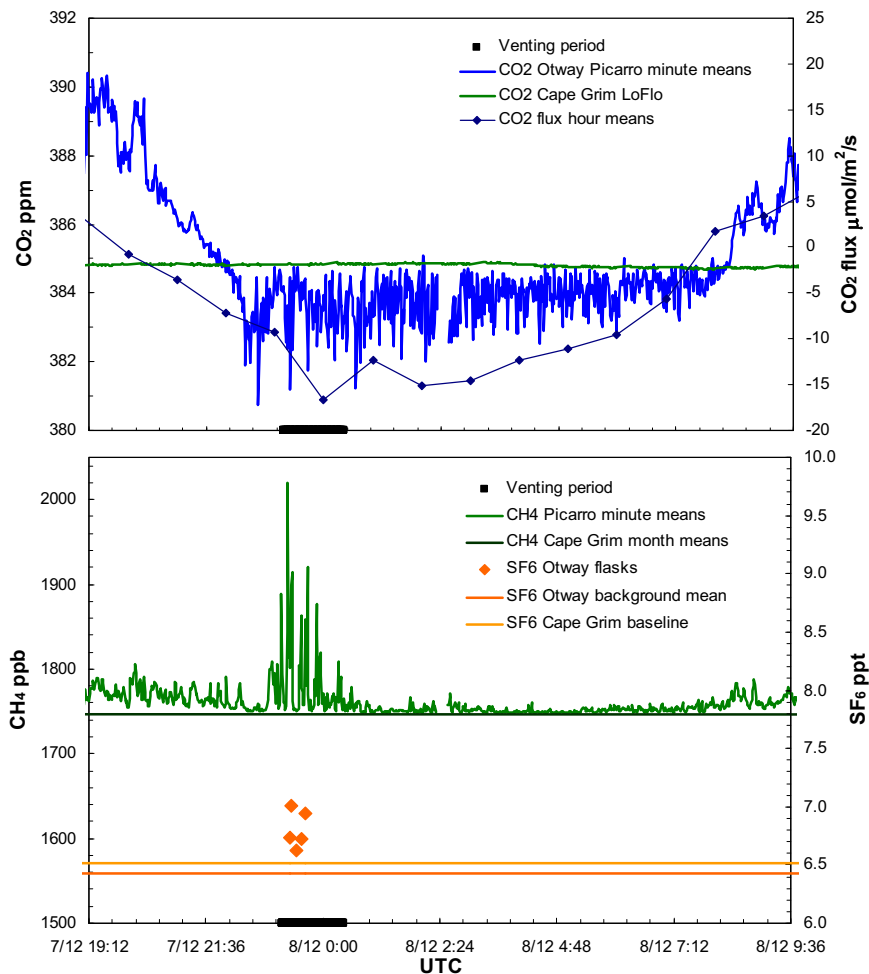


Figure 4. Atmospheric concentrations during scheduled venting of well fluid on December 8, 2009. CO₂ and CH₄ concentrations are minute mean values. Hour mean land-air fluxes of CO₂ from the EC flux tower, negative values are uptake. Flux values from before 2300 UTC were missing due to a power failure and were estimated using the mean measured fluxes for the same times over 5 subsequent days. SF₆ values are from flask sample measurements (Miller et al., 2008 [18]), each with an effective sampling period of about 3 minutes.

The dispersion of the emissions from the well was modelled using TAPM. The increases in hourly average concentrations at the atmospheric station over the period of venting were 42–45 ppb CH_4 (Figure 5), 0.015 ppm CO_2 and 0.24 ppt SF_6 , which are very similar to the mean measured increases of 33 ppb, -0.18 ppm and 0.38 ppt, respectively, compared to the following hour. Uncertainties in the modelling are estimated at about 25%, due to the hour averaging and the strong spatial concentration gradients near the measurement point (Figure 5). Peak concentration increases in the high frequency (1 minute) data during venting were about ten times higher than the hour mean. High frequency measurements greatly improve the ability to detect emissions.

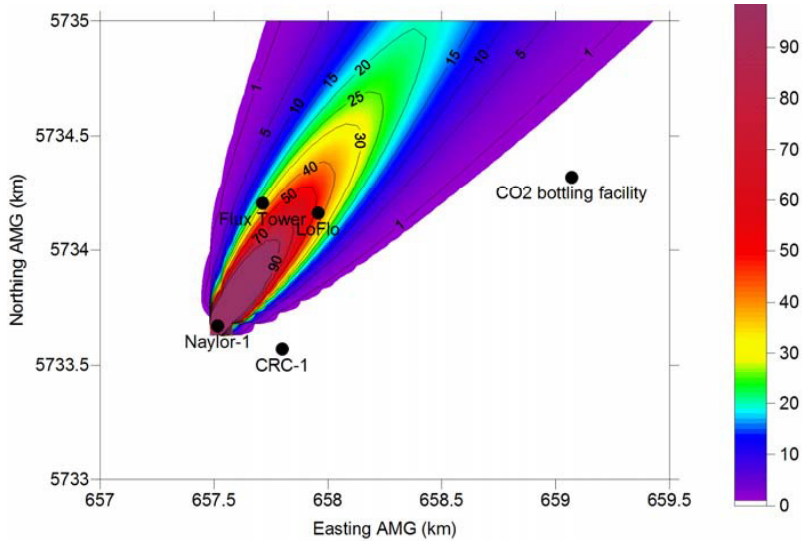


Figure 5. TAPM simulation of the CH_4 plume from the scheduled venting of the well. Shown are contours of hour mean CH_4 concentration perturbation for the period of emissions on 8 December 2009. Contours are ppb increases above background. The atmospheric monitoring location ("LoFlo"), flux tower and industrial CO_2 plant are also shown.

Emissions from the rig used to drill the CRC-1 injection well in March 2007 provide a second event to test and demonstrate the atmospheric strategy. Daytime CO₂ concentrations during south winds were within 0.5 ppm of the Cape Grim baseline, indicating minimal ecosystem activity during the dry summer period, consistent with the EC flux measurements of less than 2 μmol CO₂ m⁻² s⁻¹. When the wind shifted from south to southwest, daytime CO₂ concentrations increased by about 3 ppm. The δ¹³CO₂ signature of the extra CO₂, found using the well-known Keeling plot technique (Keeling, 1958 [19]), was -15 ‰, indicating that the source of the CO₂ was unlikely to be magmatic. Measurements of CO were about 27 ppb above background, equivalent to an emission factor of about 10 ppb CO per ppm CO₂, indicating a combustion source. These tracers, together with air back trajectories, confirm that the emissions were most likely from the engine exhausts of the drill rig and generators, independently estimated at 6 t CO₂ d⁻¹, or about twice the target leak rate.

The third event investigated was a period of unusually high nocturnal CO₂ concentrations (up to 600 ppm) in October 2008. This was originally suspected to be caused by emissions during scheduled venting from the industrial CO₂ plant. Flask air samples were collected automatically during the night time and when the wind direction was suitable. The δ¹³CO₂ value of the CO₂ source was found to be -29 ‰, consistent with the value for the local ecosystem and significantly different from the magmatic source of the CO₂ plant. Relatively low CO enhancements (0.3 ppb CO per ppm CO₂) ruled out combustion sources as the cause. Together with high measured CO₂ fluxes (between 5 and 15 μmol CO₂ m⁻² s⁻¹), these measurements confirmed that the source of the CO₂ causing the high concentrations was simply ecosystem respiration, which was highly active during spring and trapped within an extremely stable nocturnal boundary layer.

These examples show the utility of the atmospheric measurements in detecting anomalous CO₂ signals and attributing them to particular sources. Measurements of CO₂ concentrations require additional CO₂ flux information to identify leakage signals from background variations caused by the local ecosystem. However, the signals in CO₂ alone are small and tracers of the fluid help to significantly reduce the monitoring uncertainty. Measurements of tracers of CO₂ emissions that are unrelated to CCS (such as CO from combustion sources) can also reduce the risk of wrongly attributing emissions to storage leaks.

The atmospheric data obtained from monitoring CCS sites can have important uses in addition to storage leak detection. Records of the composition and fluxes of CO₂ and other gases can provide constraints on their budgets on local to continental scales, with uses ranging from agricultural management to national greenhouse gas inventories. Uncertainties in the “top down” approach of determining emissions will be reduced with more monitoring stations obtaining precise, intercalibrated and continuous records over long periods. For example, the CO₂ concentrations, fluxes and meteorological data from Otway have been used to improve the performance of the coupled TAPM-CABLE ecosystem model in simulating the ecosystem fluxes and the dispersion characteristics of that region (Luhar et al., 2009 [10]).

6. Conclusions

The Otway Project has provided a platform for the real-world implementation of an atmospheric monitoring program for onshore geological storage of CO₂. The atmospheric monitoring is designed to be sensitive enough to verify that storage of a commercial quantity of CO₂ is sufficiently leak tight to meet the requirements of sustained CO₂ emissions reduction and climate mitigation. The Otway site is in many ways a simple setting for atmospheric monitoring, being relatively flat and homogeneous and without major population centres or industries nearby. However, the ecosystem fluxes of CO₂ are very large and create a significant challenge to discriminate other sources of CO₂, including possible leakage. The experience at Otway confirms the findings from our theoretical and field experiments that monitoring CO₂ alone will be insufficient in many settings. Our approach uses a combination of techniques to overcome this challenge, including the use of selective wind trajectories, taking advantage of suitable periods in the diurnal and seasonal cycles of CO₂ and meteorology (well mixed daytime conditions during the dry season), measuring and modelling the ecosystem CO₂ fluxes, and measuring compounds that are naturally occurring or introduced tracers in the stored fluid or are indicators of CO₂ from non-storage sources. We have taken advantage of scheduled venting events at Otway, in the absence of storage leaks, to test our strategy and its sensitivity of detection. The monitoring installation could be easily and economically enhanced with measurements at several locations, rather than just the one at Otway, and with the ability to continuously measure additional tracer

compounds such as SF₆. This would improve the detection limit and the uncertainty in estimating the emission rate if leakage occurs.

A similar atmospheric monitoring strategy could be tailored to other storage sites by selecting the most appropriate techniques from the suite of concentration and flux monitoring tools (Leuning et al., 2008). While many of these tools provide continuous measurements, ongoing monitoring will also need to continuously interpret the data for evidence of leakage but without the benefit of prior knowledge of “emission” events as we have used here. Instead, we envisage a strategy involving the continuous surveillance of multiple indicators (atmospheric and subsurface measurements) in an operational framework, followed up by more intense measurement campaigns and data interpretation if an anomaly that is consistent with leakage is suggested.

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