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Atmospheric tomography as a tool for quantification of CO_2 emissions from potential surface leaks: Signal processing workflow for a low accuracy sensor array

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Abstract

Atmospheric tomography is a monitoring technique that uses an array of sampling sites and a Bayesian inversion technique to simultaneously solve for the location and magnitude of a gaseous emission. Application of the technique to date has relied on air samples being pumped over short distances to a high precision FTIR Spectrometer, which is impractical at larger scales. We have deployed a network of cheaper, less precise sensors during three recent large scale controlled CO_2 release experiments; one at the CO2CRC Ginninderra site, one at the CO2CRC Otway Site and another at the Australian Grains Free Air CO_2 Enrichment (AGFACE) facility in Horsham, Victoria. The purpose of these deployments was to assess whether an array of independently powered, less precise, less accurate sensors could collect data of sufficient quality to enable application of the atmospheric tomography technique. With careful data manipulation a signal suitable for an inversion study can be seen. A signal processing workflow based on results obtained from the atmospheric array deployed at the CO2CRC Otway experiment is presented.

Keywords: carbon dioxide; geological storage; geosequestration; monitoring; atmospheric tomography; Bayesian inversion

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1. Introduction

Having techniques available for the accurate quantification of potential CO_2 surface leaks from geological storage sites is critical for regulators, public assurance and for underpinning carbon pricing mechanisms. Currently, there are few options available that enable accurate CO_2 quantification of potential leaks at the soil-atmosphere interface. Integrated soil flux measurements can be used to quantify CO_2 emission rates from the soil [1] and atmospheric techniques such as eddy covariance [2], atmospheric transport modelling [3] or Lagrangian stochastic modelling [4] have been used with some success to quantify CO_2 emissions into the atmosphere from controlled CO_2 surface emissions.

A new technique to quantify CO_2 emissions was trialed at the CO2CRC Ginninderra controlled release site in Canberra [5]. The technique, termed atmospheric tomography, used an array of sampling sites and a Bayesian inversion technique to accurately and simultaneously solve for the location and magnitude of a simulated CO_2 leak. The technique requires knowledge of concentration enhancement downwind of the source and the normalized, three-dimensional distribution (shape) of concentration in the dispersion plume. Continuous measurements of turbulent wind and temperature statistics were used to model the dispersion plume.

The first demonstration of the atmospheric tomography proved successful. CO_2 was released at a rate of 82 kg/d from a surface source located within an array of eight evenly spaced sampling points on a 20 m radius circle. Air samples were pumped to a high precision Fourier Transform Infrared (FTIR) Spectrometer and analysed sequentially for atmospheric CO_2 concentrations (8 sample points per 30 mins). The emission rate was determined to within 3% of the actual release rate and the localisation within 4% of the correct position [5]. Applying the technique at a larger scale, however, requires a more practical configuration, using multiple, low cost sensors, but this will be at the cost of data quality. Therefore a network of eight portable CO_2 sensors was developed and debugged during this experiment and then deployed on two other sites. This paper assesses the option of using these multiple, comparatively inexpensive CO_2 sensors, but with less precision and accuracy. It assesses whether a useable signal can be extracted from such datasets for the application of atmospheric tomography.

2. Experimental description

2.1. Site descriptions

The CO2CRC Otway Project, located in southeast Australia (38°31'50''S, 142°48'42''E, 50 m elevation), is a midscale geological storage demonstration project utilizing a depleted gas field for storage [3] (Figure 1a). During Stage 1 of the Otway Project, some 65,445 t of CO_2 rich gas was stored at the site from March 2008 to August 2009. In 2011, the Otway Stage 2B residual saturation and dissolution test was held. It was the first field test of its kind to measure large-scale residual trapping of CO_2 within a field project [6]. A byproduct of the field test was the controlled release of CO_2 during the water lift and well venting activities. This provided a source of CO_2, which was used to field test the atmospheric tomography technique. The equivalent of approximately 3 t of CO_2 was released per day over the period of July-September 2011. Releases occurred on 23 days during this time. On occasions, the equivalent emitted CO_2 flow rate was 9-15 t/d but only for short durations (typically 1-8 hours).

Immediately after the CO2CRC Otway experiment, the CO_2 monitoring equipment for the atmospheric experiment was relocated to the Victorian Government Department of Primary Industries, Australian Grains Free Air CO_2 Enrichment (AGFACE) facility in Horsham, Victoria (36°45’07’’S, 142°06’52’’E, 127m elevation). At the AGFACE facility, the 8 independent sensors were deployed on the perimeter of a 7.5 hectare mixed agriculture crop site (wheat and field peas) and monitored the site from October-November 2011 (Figure 1b). There are eight AGFACE rings (16m diameter each) located within the field and CO_2 is released into the atmosphere on the upwind side of the ring, through 0.3 mm holes (20 holes/m) drilled by laser on 23 mm i.d. stainless steel tubes at supply line pressures up to 500 kPa and raised approximately 1m above the ground surface [7]. The total amount of CO_2 released (only during daylight hours) at the AGFACE facility from 13 October to 28 November 2011 was 303 t, equating to approximately 4 t/d.

2.2. CO_2 towers and 3D sonic anemometers

The same set of autonomous CO_2 sensors was used at both the CO2CRC Otway site and AGFACE facility. Each CO_2 tower was equipped with a Vaisala CARBOCAP GMP343 CO_2 sensor, Vaisala HMP60 relative humidity and temperature sensor, a 55 W solar panel, 60 Ah deep cycle battery, antenna, Campbell Scientific CR1000 data logger and Maxon Modmax 3G modem (Figure 2). The CO_2 sensors were positioned approximately 2 m above the ground surface at each location. For the field trial at the CO2CRC Otway site, the eight towers were located around the CRC2 well at distances ranging from 154 to 473 m from the well. One minute averaged CO_2 concentration data was collected every minute and downloaded remotely every few days. A 3D sonic anemometer (Type HS, Gill Instruments Ltd) located on an eddy covariance tower, co-located at CO_2 tower number 6, at the CO2CRC Otway site provided the x, y, and z components of the wind vector at 20Hz. These data provided the Monin−Obukhov stability parameter (MO length), friction velocity, u*, mean horizontal wind speed, U, and wind direction, θ. These quantities are required for the atmospheric tomography and model the shape and direction of the dispersion plume downwind of the source [5]. For the AGFACE site, an eddy covariance instrument was installed in the centre of the experimental field at 2 m height for the duration of the experiment and provided 3D wind speed and direction data, as well as boundary layer parameters.

2.3. Limitations with the GMP343s

The CO2CRC has been assessing the performance of the GMP343 sensors over many years [8]. In spring 2010, data were gathered from eight co-located FTIR inlets and the GMP343 sensors at the Ginninderra controlled release facility over a period of 3 months and this enabled a more detailed evaluation of their performance (for detailed description of the experiment setup, refer to Humphries et al [5]). The FTIR provided on-the-fly calibration for the GMP343 sensors. It was decided not to calibrate the GMP343 sensors regularly, but to investigate their behaviour over the three months by comparison with the FTIR. Applying empirical corrections for drift and temperature, the overall scatter in the residuals could be reduced to the 2-4 ppm range. Hence the sensors are reasonably precise. Given these limitations, relying on the sensors to provide absolute CO_2 concentration measurements due to a CO_2 release would be difficult (unlike other studies using more accurate instrumentation, c.f. Jenkins et al. [3]); however, because the sensors are reasonably precise, perturbations above a relative signal may still prove useful. Locating a source depends on relative, not absolute signals, as explained in Humphries et al. [5].

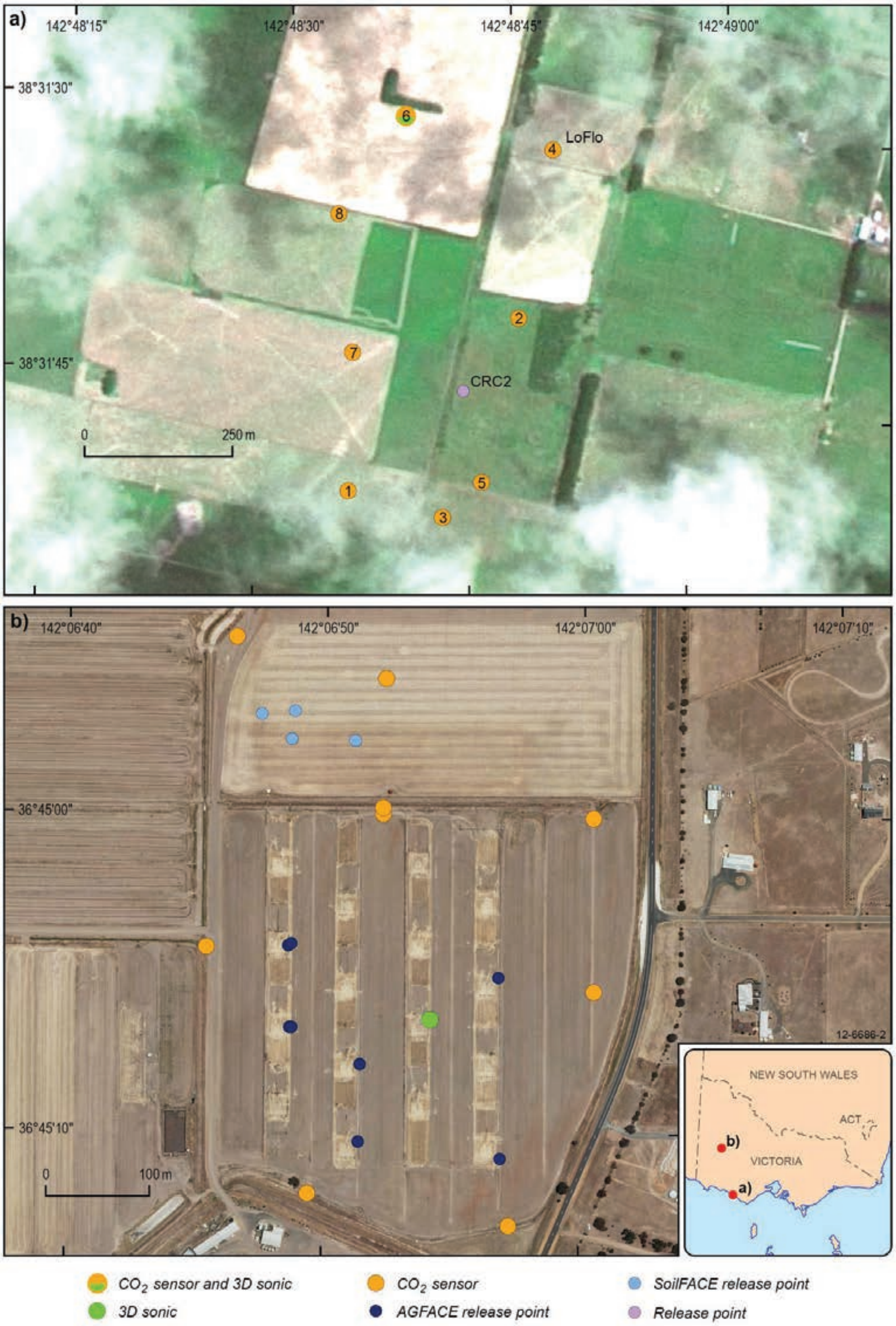

Figure 1. CO_2 sensor arrays deployed at the a) CO2CRC Otway site and b) AGFACE facility

Figure 2. Photo of one of the eight CO_2 sensors at the AGFACE facility. The GMP343 is located at the top of the tower.

3. Workflow for CO_2 data processing

The software package Mathematica (Wolfram) was used to manipulate the large volumes of data (over 100,000 entries per sensor). Concentration data were available at 1 minute intervals from 30 June 2011 to 10 October 2011. The individual steps for data processing are outlined below and a summary provided in Figure 3:

1. **Starting with the 'raw' ppm data:** The raw data available for analysis have limited automatic correction factors applied (relative humidity, pressure and temperature) according to manufacturer's specifications. Refer to Figure 3 for the workflow of applying corrections. It is important to ensure the software being used recognises the date and time format and that all date-strings are correct and consistent. This can be a very time consuming step during data processing.
2. **NumericQ:** Only data were used where all eight CO_2 sensors had concentration values recorded. Some data drop-outs resulted in some missing data from some sensors. Due to the nature of the Bayesian tomography application, data from all eight CO_2 sensors are required to generate an accurate result. It is also important to ensure any Null, -9999 or zero/negative values are removed where data drop-outs occurred.
3. **Baseline trend removal:** Each CO_2 sensor displayed some minor drift over the measurement period. This baseline trend was removed by finding the running minimum over a three day period and subtracting the resulting linear trend from the concentration data. This removes sensor drift errors that could be caused by temperature, pressure or humidity variations for the duration of the measurement period.

4. **Calculating a CO_2 background for the site:** An overall (baseline minimum subtracted) background concentration was calculated for the overall site by averaging the eight CO_2 sensor concentration values with the sensor drift trend removed as described in step 3.
5. **CO_2 concentration above background (calculating the CO_2 perturbation):** For each sensor, the 'true' CO_2 concentration above background was then calculated. This involved processing the concentration data up to step 3 mentioned above, then removing the calculated site 'background' (step 4) to result in a CO_2 concentration above or below natural background levels.
6. **Comparing concentrations with wind data:** Before the CO_2 perturbations can be compared to wind direction some processing of the wind components is required. Wind data were available from a sonic anemometer in 15 minute readings. This meant that in order to compare wind with the CO_2 perturbation, an interpolation of wind data was required to simulate 1 minute wind direction values. After this processing step, it is then possible to compare processed CO_2 perturbations with wind direction.
7. **Assessing conditions of interest:** It was now possible to compare and assess CO_2 perturbations with wind direction to see if higher values can be seen from the wind direction of a known CO_2 release. It may be useful to assess all data compared to data during known release times only, or day-time perturbations only. Finding the average perturbation for each 5° or 10° sector and plotting the error associated with this can also be extremely useful for data analysis.

These are the processing steps used for the preliminary assessment of the Otway Stage 2B controlled release experiment. The assumption that the wind data can be interpolated over one minute intervals from 15 min data is unlikely to hold under low wind speed conditions. Under such conditions, the wind direction may swing widely over a 15 min period. This error will be minimized in the later tomography analysis by filtering out data with the friction velocity parameter $u^* \leq 0.15$ m/s since the Monin−Obukhov theory for dispersion is unreliable under light wind conditions.

Several of the steps outlined above can be improved, particularly for calculation of the background CO_2 [5]. Another processing step that could be applied to improve the data quality and reliability includes application of more refined temperature correction for each of the sensors. It was observed during the first tomography demonstration project that the CO_2 sensors appear to have a sensitivity to higher temperatures (e.g. >30 °C) which the manufacturer corrections do not account for. Some negative data glitches are also still under investigation.

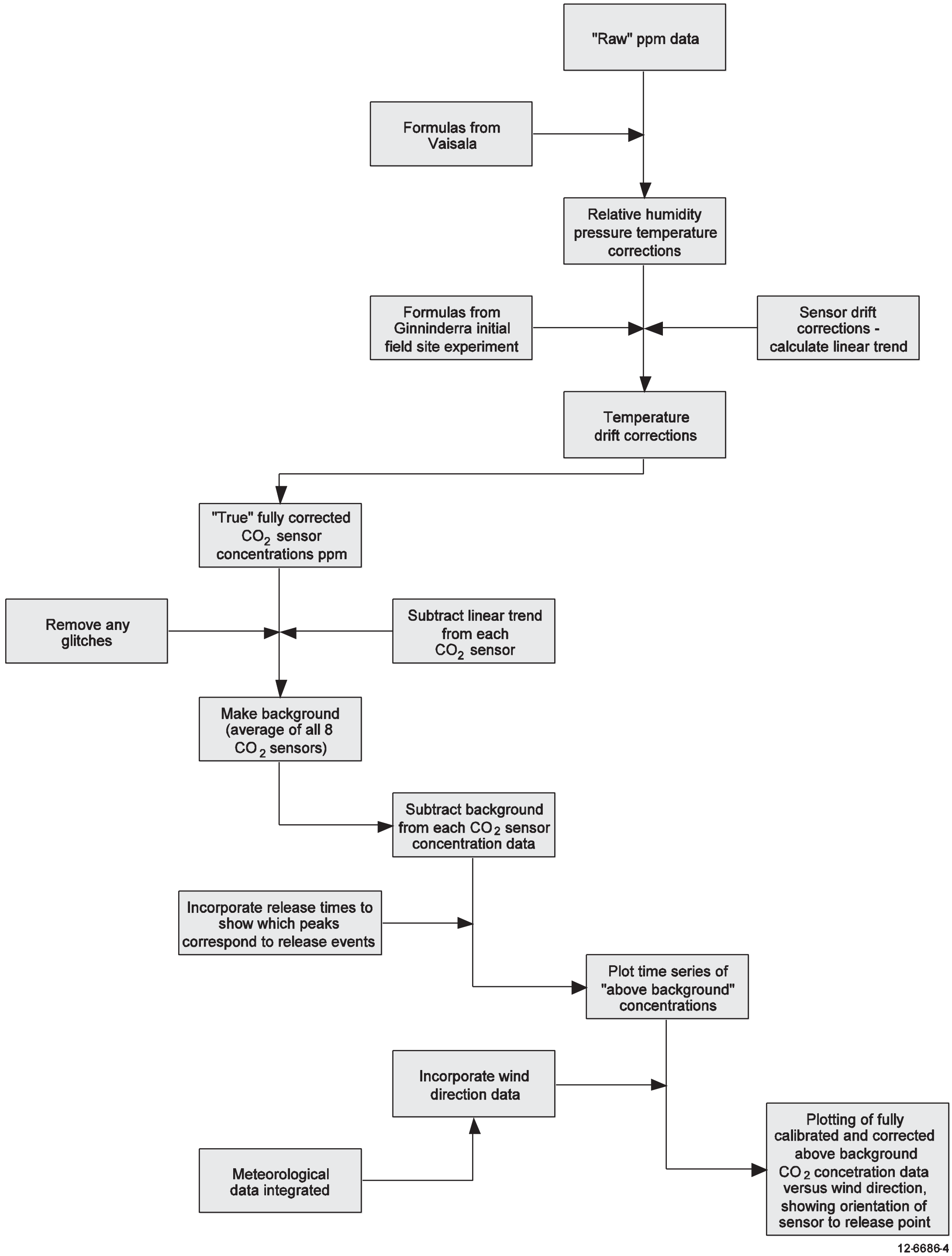

Figure 3. Flowchart for data processing

4. Results

4.1. CO2CRC Otway case study

Figure 4 provides an illustration of the significant inter-day variability of the CO_2 measurements at the Otway site and represents the averaged minute CO_2 concentration from all eight CO_2 sensors (baseline minimum subtracted). Natural variations in CO_2 of up to 200 ppm above the baseline minimum (approximately 360 ppm) are observed and these are predominantly due to a combination of nighttime plant respiration and stable atmospheric inversions [9]. Also indicated on the plot are the times when CO_2 was released from the CRC2 well. Figure 4 was used as the background dataset for the array and a CO_2 perturbation plot for each sensor was derived by subtracting this background plot from each concentration minus the baseline sensor plot. Figure 5 shows the dominant wind directions for the duration of the measurement period with the bearings of each sensor shown to show which sensors are expected to show CO_2 perturbations. The results of these subtractions, plotted versus wind direction, are given in Figure 6.

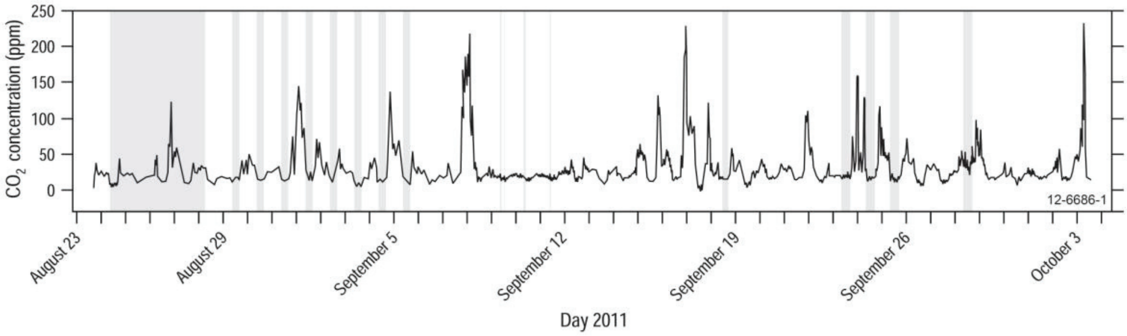

Figure 4. Average background concentration for all eight sensors (baseline minimum subtracted) and CO_2 release periods indicated in shaded areas.

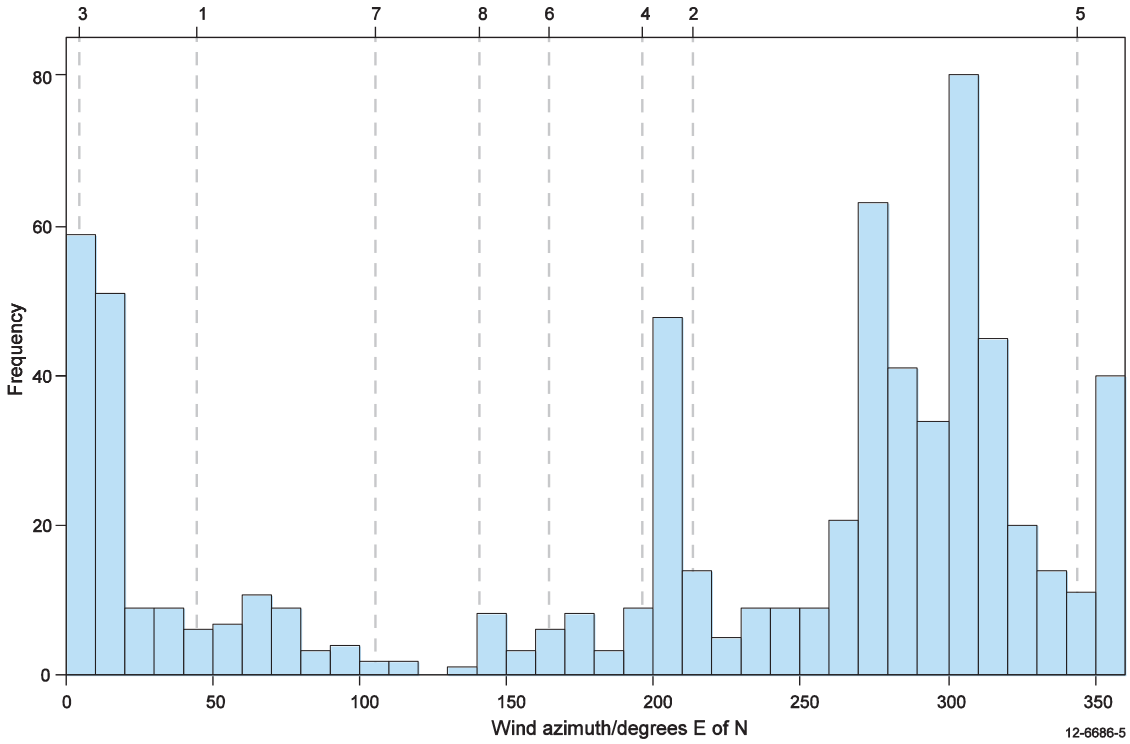

Figure 5. Frequency of wind direction for the duration of the Otway Stage 2B release periods with sensor bearings indicated as grey vertical dashed lines.

Despite the large variability in CO_2 concentrations, the simple data processing procedure outlined above does extract useful data. This is because the technique is essentially differential, using the differences between sensors and not the absolute values. Perturbations can be clearly observed for CO_2 sensors 1, 2, 5 and 7 in Figure 6 and these largely coincide with the anticipated wind directions based on the bearing of the sensors from the CRC2 release well. No perturbations are evident for CO_2 sensor 8. A small perturbation is observed for CO_2 sensor 3 and no perturbation is observed for CO_2 sensors 4 and 6 (data not shown). While CO_2 sensors 2 and 5 are approximately 150 m from the release point, CO_2 sensors 1 and 7 are 256 m and 197 m distant, respectively. Perturbations were not observed at the most distant CO_2 sensors (4, 6 and 8), although these were all located in a northerly direction from the CRC2 well where the frequency of wind direction for this sector during release times is low (Figure 5). The peaks from three of the sensors, if propagated linearly back along the wind direction, define a triangular region about 20 m on a side within 20 m of the actual site of the release. The fuller Bayesian inversion, including dispersion modelling, will refine this estimate and provide better uncertainties. Small signals from other sensors, currently not apparent, may also further constrain the position when a full model of the dispersion is used. The signal at Sensor 7 seems to have originated at a nearby CO_2 processing plant, about 1 km distant; emissions from this plant have been detected on other occasions. As explained in Humphries et al [5], the tomography technique works well when data can be accumulated over a range of wind directions, which may take several weeks depending on site-specific wind patterns. Table 1 summarises the CO_2 sensor bearings and distances from the CRC2 well.

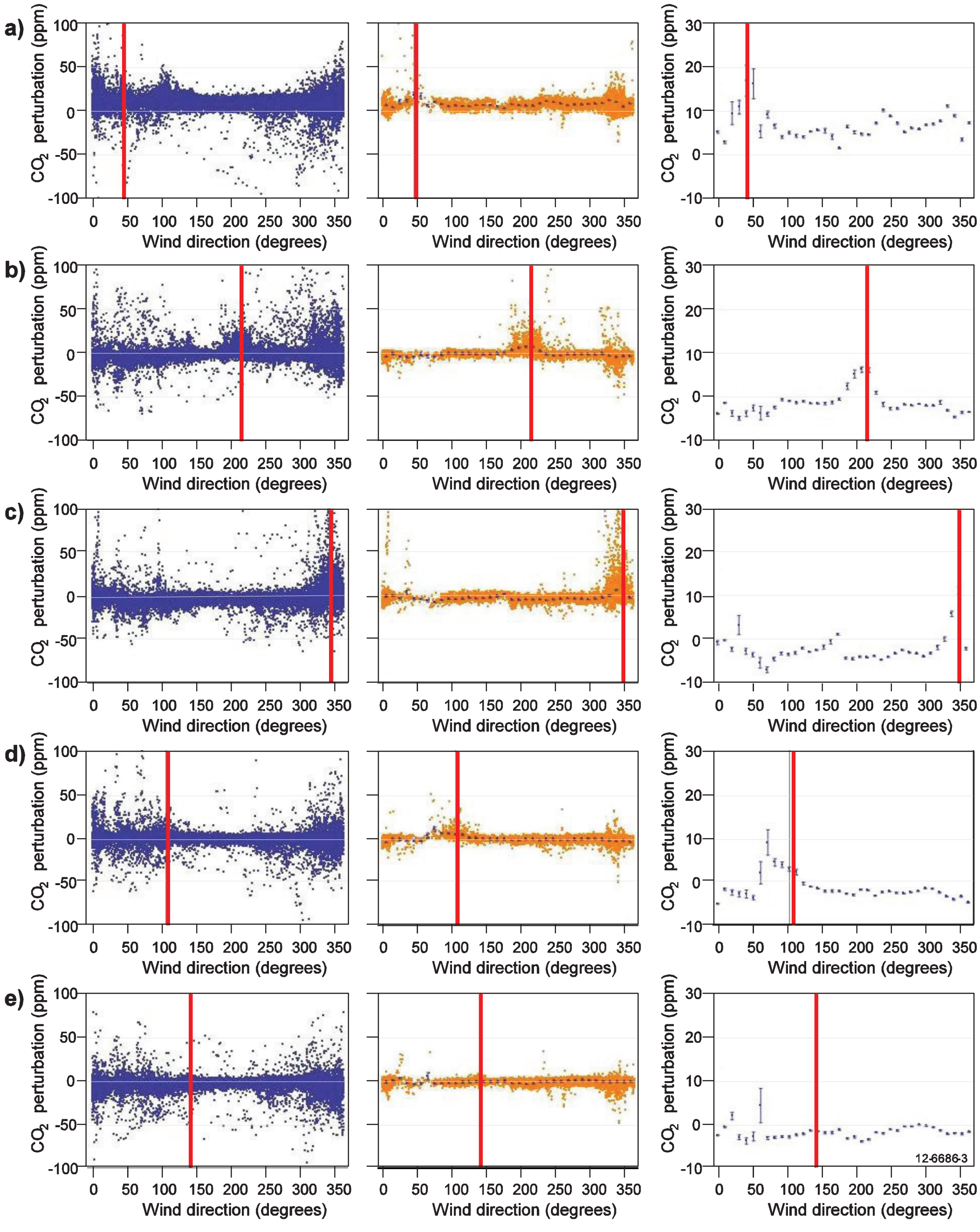

Figure 6. Otway site: Example of CO_2 sensor results a) sensor 1; b) sensor 2; c) sensor 5; d) sensor 7; and e) sensor 8. Left) CO_2 perturbations for all data against wind direction; middle) CO_2 perturbations during release periods against wind direction overlaid with mean CO_2 perturbations; right) Mean CO_2 perturbations for each 10° wind direction 'bin' and corresponding standard errors of the mean against wind direction. Vertical red lines indicate bearing from the CRC2 well CO_2 release point.

Table 1. CO_2 sensor bearings and distances from the CRC2 well (CO2CRC Otway site)

CO_2 Sensor #	Distance to CRC2 release well (m)	Bearing to CRC2 release well (°)
1	256	45.0
2	154	213.1
3	215	5.3
4	433	196.3
5	156	344.7
6	473	164.2
7	197	105.4
8	364	141.1

The data in Figure 6 are preliminary, but the clear detection of the concentration enhancements at the appropriate wind directions shows that a useful, simply geometric estimate could be made of the location of the source. Including dispersion modelling and the Bayesian inversion will improve this somewhat and allow an estimate of average release rate to be made, together with error estimates. This work is in progress.

4.2. AGFACE study

Installation of the sensors on yet a larger scale at the AGFACE facility in a more isolated region introduced new challenges. Remote access to the sensors was intermittent and it was not possible to log remotely into one of the sensors. Data for this sensor required manual download every 7-10 days. Two months of data were collected during the trial and these will be processed as outlined in the CO2CRC Otway case study above. The AGFACE dataset represents a greater challenge for application of the atmospheric technique due to the presence of multiple, somewhat extended sources. It is not clear at this stage whether there will be perturbations of sufficient magnitude suitable for the application of atmospheric tomography, but this is the subject of ongoing work.

5. Discussion

The advantage of atmospheric tomography is that it can solve for the location and source strength of a simulated CO_2 leak simultaneously, and this will be the next step in our data processing and modelling. In the absence of that analysis, a geometric estimate of the location can be made based on the maximum observed perturbations where there is a clear peak around a wind direction, i.e. 45° for Carbocap 1, 205° for Carbocap 2 and 345° for Carbocap 5. Using these results to estimate a CO_2 source using wind direction where a large perturbation was detected and finding where several sensors 'cross-over' using this technique will give a reasonable estimate to the location of a potential CO_2 leak. The error would be indicated by the degree by which each sensor line does not quite cross at the same point. Even rough positional estimates such as these would be useful for CCS monitoring and detection because leak detection is hierarchical. Once it has been identified that a leak exists, more detailed monitoring in the suspected area would need to take place.

6. Conclusion

After initial processing of the atmospheric CO_2 concentration data collected during the Otway Stage 2B project, preliminary analyses show promising results for CO_2 perturbations above background during favourable wind directions. Some corrections still need to be applied to the data to improve data precision and finding data during periods of interest will improve detection of perturbations. We anticipate the data will be suitable for the next stage of our work and application of more elaborate modelling and atmospheric tomography.

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References

[1] Lewicki JL, Hilley GE, Dobeck L, Spangler L. Dynamics of CO_2 fluxes and concentration during a shallow subsurface CO_2 release. *Environ Earth Sci* 2010; **60**:285-297.

[2] Lewicki JL, Hilley GE. Eddy covariance mapping and quantification of surface CO_2 leakage fluxes. *Geophys Res Lett* 2009; **36**: L21802.

[3] Jenkins CR, Cook PJ, Ennis-King J, Undershultz J, Boreham C, Dance T, et al. Safe storage and effective monitoring of CO_2 in depleted gas fields. *Proc National Acad Sci* 2012; **109**:E35-E41.

[4] Loh ZM, Leuning R, Zegelin SJ, Etheridge DM, Bai M, Naylor T, et al. Testing Lagrangian atmospheric dispersion modelling to monitor CO_2 and CH_4 leakage from Geosequestration. *Atmos Environ* 2009; **43**:2602-2611.

[5] Humphries R, Jenkins C, Leuning R, Zegelin S, Griffith D, Caldow C, et al. Atmospheric Tomography: A Bayesian Inversion Technique for Determining the Rate and Location of Fugitive Emissions, *Environ Sci Technol* 2012; **46**:1739–1746.

[6] Paterson L, Boreham C, Bunch M, Dance T, Ennis-King J, Freifeld B, et al. Overview of the CO2CRC Otway residual saturation and dissolution test. *Energy Procedia* 2012 (in press)

[7] Mollah MR, Norton RM, Huzzey J. Australian grains free air carbon dioxide enrichment (AGFACE) facility: design and performance, *Crop & Pasture Science* 2009; 60, 697-707.

[8] Bernardo C, De Vries DF. Permanent shallow subsoil CO_2 flux chambers for monitoring of onshore CO_2 geological storage sites, *International Journal of Greenhouse Gas Control* 2011, **5/3**: 565-570.

[9] Etheridge D, Luhar A, Loh Z, Leuning R, Spencer D, Steele P, et al. Atmospheric monitoring of the CO2CRC Otway Project and lessons for large scale CO_2 storage projects, *Energy Procedia* 2011; **4**:3666–3675.