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A new technique for monitoring the atmosphere above onshore carbon storage projects that can estimate the locations and mass emission rates of detected sources

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Abstract

Carbon Capture and Storage (CCS) projects could grow to become a major new industrial activity over the next few decades; but securing the associated climate benefit is critically dependent on ensuring high integrity containment of the injected CO_2. Our technique, called LightSource, is based on commercially available optical gas sensors that measure path-averaged CO_2 gas concentrations along beams scanned over part of an onshore CCS site. Inter-beam correlations are used to infer the current local ambient background concentration. Statistically significant discrepancies between the multiple beams' path-averaged concentration measurements can be used to infer the existence of a source by applying the methods of statistical process control. This allows the estimation of the anomalous concentration on each beam that is associated with the inferred source(s). Using these anomalous concentration data in conjunction with a gas dispersion model, high frequency wind velocity and turbulence intensity data, we can solve the inverse gas dispersion problem to estimate the location and mass emission rates of the source(s) that best explain the data. The system does not require sources to be situated within the beam pattern unlike tomographic approaches which in addition require more intensive instrumentation. We have evaluated the LightSource system's performance under field conditions at the Quest CCS project site in Alberta Canada. All calibrated releases of tempered CO_2 at emission rates of up to 300 kg/hr were successfully detected.

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1. Introduction

We will set the technology background by reviewing the techniques normally proposed to monitor the atmosphere above CCS projects for any signs of CO_2 emissions and explain why we consider none of the existing methods to be adequate. We then describe a new technique we have invented called LightSource, which offers the prospect of continuous, automated, large-area monitoring of onshore CCS projects at a sensitivity sufficient for detecting sources capable of significantly jeopardizing containment targets. This method utilizes a Gaussian plume dispersion model as discussed in Hirst et al. [1]. Using anomalous CO_2 concentration data, high frequency wind velocity and turbulence intensity data, the inverse gas dispersion problem is solved to estimate source locations and mass emission rates. We will also present some of the experimental results for the estimated sources' locations and their mass emission rates from a controlled release test performed at one of the three injection pads at the Quest CCS project site. Quest is the first Shell-operated Carbon Capture and Storage (CCS) project with a planned injection rate of 1.1 million tons of CO_2 per year for a total of 25 years. The storage operation commenced in August 2015 and has already safely stored over 1 million tons of CO_2 in a deep saline aquifer. Our system successfully detected all controlled releases and provided estimates for the locations and mass emission rates of the sources responsible. Finally, we will critically evaluate the performance of this first LightSource installation and describe improvements for future implementations.

1.1. Technology background

There are no established techniques suited to the detection and quantification of gas emissions from onshore carbon dioxide CO_2 sequestration sites. A system that can continuously and reliably perform this task will enhance the safety case and contribute to the containment verification of sequestration projects. The usually proposed approaches to achieving this are: simple point gas detectors, eddy covariance methods and gas flux chambers. More recently, tomographic approaches to atmospheric detection of carbon dioxide leaks have been proposed by Levine et al. [2]. Such approaches require substantially denser instrumentation effort to provide a "net" of sensor beams spanning the area of interest; but cannot locate sources outside of the beam array as LightSource can.

1.1.1. Point gas sensors

Gas sensors typically measure the concentration of gas at some specified point, and that concentration is usually stated in parts per volume, ppv. When using a single point gas sensor one might detect an elevation of gas concentration resulting from a leak elsewhere – but only if the gas sensor is positioned within the plume of dispersing gas, is sufficiently sensitive and can respond in an appropriate time. If the wind direction takes the gas away from the sensor the leak will not be detected. Even if the sensor is within the dispersion plume, we have no information as to where the plume is situated in relation to the gas sensor: are we on the edge or near the plume center, close to the source or far away? We know there is a gas leak, but we do not know if the leak is a modest leak a few meters upwind or a catastrophic leak several km away. In short, concentration measurements cannot be treated as a proxy for mass release rate. However, if one deploys an extensive array of suitable sensors then one can discern concentration field gradients and begin to approximately locate the leak's source; but only if all the sensors are sufficiently precise and calibrated to reflect real changes in concentration within the gas plume and offsets between individual sensors' calibrations are small in relation to the changes in concentrations measured. In addition to these limitations, the cost and maintenance of such a system would be prohibitive.

1.1.2. Eddy covariance or eddy correlation techniques

Eddy covariance or eddy correlation techniques are an advanced technique developed to measure gas fluxes remotely; they are widely used in environmental studies to characterize the emissions from generic land types, e.g.: swamps, rice-paddies, grasslands, permafrost, forests etc. The method comprises making rapid simultaneous measurements of wind velocity and gas concentration. Both measurements need to be performed at high data rates and with high precision; the associated specialist sensors are typically mounted on a tall mast or a tower. The underlying principle is that over a suitably chosen period of time there will be no net mass transport of air vertically. Thus if one multiplies the instantaneous gas concentration data by the corresponding vertical component of wind velocity one can discern whether there is a net vertical transport of the gas species of interest: if air travelling upwards has a greater density of CO_2 than air travelling downwards then there is a upward flux of CO_2. In this way a representative mass flux rate for the gas species of interest can be obtained for the instrument's setting. However, the flux value obtained relates only to a certain footprint area. As the wind direction changes the footprint shifts around correspondingly. More significantly though, as the solar insolation, wind-speed, turbulence, atmospheric boundary layer depth, etc. change so does the shape, size and range of the associated footprint area. For this reason the method is only suited to providing a representative flux value characterizing a homogeneously emitting source area present upwind of the tower; it is not suitable for mapping inhomogeneous source fields: i.e. discrete sources!

1.1.3. Flux chamber methods

Flux chambers are typically flattish and cylindrically shaped containers; they are open at the base and when placed on the ground gas migrates into the chamber, increasing the gas's concentration. Knowing the base area, the volume of the container and the rate of increase in gas concentration, the ground surface's mass flux rate can be inferred. This is the so-called passive flux chamber or accumulation chamber: the gas just accumulates with concentration increasing in time. A variant of the method is called a dynamic, or active, gas flux chamber; in this approach a diluent gas known to be completely free of the gas species of interest is introduced into the chamber at a constant volume flow rate. The chamber has a vent that allows excess gas to escape without over-pressuring the chamber. Sample gas at a sufficiently slow volume flow rate is extracted and its concentration measured and discarded from the system. Over time the gas of interest's concentration increases asymptotically towards a dynamic equilibrium value. The mass in-flow of gas through the base is then exactly equal to the mass necessary to bring the volume flow of the diluent gas up to the equilibrium concentration. The flux rate is then obtained by using the concentration, the area of the chamber's base and the diluent's volume flow rate. While potentially providing some useful information on gas flux rates, flux chambers are unsuited to continuously monitoring the full area of the sequestration site; they are incapable of detecting or locating emission hotspots and cannot provide long-term trend data without a continuous, long-term, labor intensive measurement campaign persisting throughout the containment verification program.

2. Discussion

We have previously successfully demonstrated the LightSource method using methane but its use for CO_2 is considerably more demanding for two reasons: the natural globally averaged background concentration of CO_2 is about 400 ppm (versus 1.8 ppm for methane) and can vary by more than a 100 ppm within hours (methane's corresponding variability is 10-20 ppb). This extreme CO_2 background variability is due to local, extensive diffuse sources and sinks associated with photosynthesis and respiration. This makes it doubly difficult to detect a source as the associated local concentration enhancement per unit mass released is small compared to the background concentration's dominating contribution to the path-averaged concentration. Furthermore, the variability of the background concentration complicates estimation of the precise ambient background contribution that needs to be subtracted from a measurement to reveal the local "anomalous" extra concentration associated with the local sources of interest. Therefore, there are two factors constraining estimates of anomalous concentrations: the inherent sensor precision and the associated uncertainly in estimating the true background concentration.

Since 2009 we have been cooperating with Boreal Laser Inc. of Edmonton, Alberta to secure a fundamental re-design and upgrade of their GasFinder2 open-path gas sensor. Although it was the best commercially available CO_2 beam sensor, our simulation modelling and field test data, showed it was inadequately precise for the Quest application. The new GasFinder3 system has improved sensor precision by more than an order of magnitude and reduced thermal and optical drift by incorporating advanced optical source & detection technologies and digital signal processing. This new GasFinder3 sensor was installed at Quest. The sensor's improved thermal stability and data precision made it feasible to collect and analyze data prior to first injection in August 2015 so as to better establish the natural variability of the atmospheric background concentration of CO_2. In order to evaluate the method's actual performance in the field, we arranged an extensive series of controlled releases of CO_2 at the Quest pad 8/19 to follow on from two weeks of just monitoring background concentration with the new sensor.

2.1. Experimental goals and methods

The purpose of the controlled release tests was to provide multiple data sets of path-averaged concentration data along three beams, collected using the new Boreal GasFinder3 scanned open-path gas sensor. The gas releases needed to be at ambient pressure and temperature, so as to closely replicate the anticipated behavior of a ground level area source: cold gas would disperse differently to ambient temperature gas due to its increased density and pressure-driven releases would be additionally diluted by turbulence related to the released gas's momentum. The releases also needed to be at a stable, calibrated mass emission rate, from well-defined locations.

Bearing in mind the goal was to evaluate our ability to detect, locate and quantify the source emission rates from the data, it was essential that each data set included measurements corresponding to a variety of wind conditions for each controlled release. Injection of 1.1 million tons (Mt) of CO_2 per year for 25 years means a total 27.5 Mt stored, for which 1 % loss per 100 years would correspond to a 314 kg/hr average emission rate. Hence we decided to use a 300 kg/hr release rate as our standard and make the releases from an area source, not a point source, so as to minimize the mixing effects of the residual gas flow momentum from the numerous, small, orifices used.
In the event of a real release, data would accumulate over many days and would comprise contributions for a wide variety of wind directions, speeds and atmospheric turbulence intensities – all such variability would serve to constrain the source location. However, given a mass release rate of 300 kg/hr, it was not practical to sustain a release for several weeks, which is what one would ideally have sought to do. Our experimental program was limited to five days of which the first and last were mobilization and demobilization days: leaving just three days for experiments. Given these time constraints and unpredictability of the winds, we decided to choose five release locations within the pad and standardize the emission rate and the area of the sources. Rather than release gas and wait for wind changes, we opted to standardize on a duration of 30 minutes for each release, and then relocate the release point, wait a minimum of 15 minutes for residual gases from the preceding location to clear from the site, and then start a new release at a new location but with similar wind conditions. In this way we could subsequently compile concatenated data sets for each of the locations, including data for a variety of those winds occurring during the three experimental days. By distributing the five emission locations across the pad we were able to ensure that provided the wind changed we would get data at some locations that included a variety of different beam intersections produced by different wind conditions.

2.2. Experimental facilities

Air Liquide were commissioned to design and built a rig to provide a stable, specified mass flow of tempered CO_2 gas via flexible hoses to any location within the pad boundary. Figure 1 shows a panoramic view of the approximately 100 x 100 m pad, which is enclosed by a secure chain-link fence. The small hut seen just right of center houses the power and communications equipment, the structure visible in the bottom left hand corner is the eddy covariance rig, installed and run the by University of British Columbia team. Black et al. [3] have measured naturally occurring CO_2 fluxes on the pad since June 2014.

Fig.1. Panorama of the Quest pad 8/19 near Edmonton, Alberta.

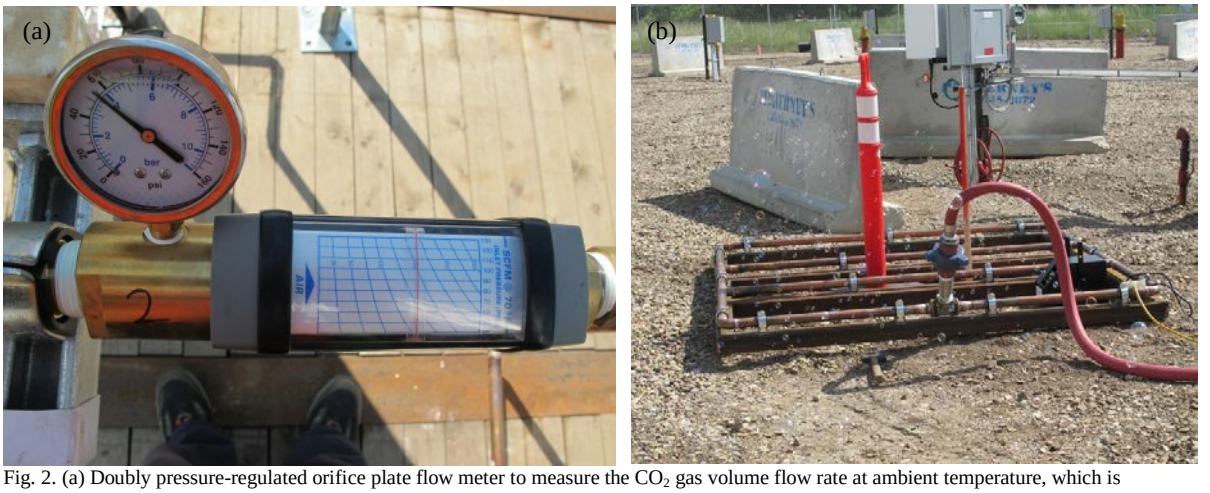

Fig. 2. (a) Doubly pressure-regulated orifice plate flow meter to measure the CO_2 gas volume flow rate at ambient temperature, which is converted using the drive pressure and density to obtain the mass release rate; (b) Release manifold with bubbles for visualization.

Gas flowed via a doubly pressure-regulated orifice plate flow meter as seen in Figure 2 (a) through 200 ft of fire-hose to the release manifold, which provided a diffuse 2 x 2 m area source of gas, released to atmosphere via numerous small holes in the copper pipework: so as to avoid any significant momentum transfer influencing the dispersion process. This better reflects the anticipated behavior of gas gently escaping into the atmosphere from the ground. A simple bubble machine provided useful visualization (see Figure 2 (b)) of the atmospheric dispersion process, the bubbles are passively dispersed, being carried by the air in the same way as low concentration CO_2 at ambient temperature would be.

Figure 3 (a) shows the Boreal Laser GasFinder3 sensor mounted in the SW corner of pad 8/19 which is automatically steered to each of three retro-reflecting panels (Figure 3 (b)) in the other corners of the square pad. The optical absorbance, measured at a characteristic CO_2 Infra-Red absorption feature wavelength, allows the total path-integrated mass concentration along the whole beam to be measured: this is a much richer, more stable and better statistical representation of how much gas is in the atmosphere than can be obtained with traditional point gas sensors. Data from the Boreal Laser GasFinder3 sensor is collected sequentially from the three beams: Beam 1 is the South-North beam, Beam 2 is the North East beam and Beam 3 is the West-East beam. The beam lengths are approximately 87 m, 125 m and 93 m respectively. The data are uploaded automatically to a Shell FTP site along with extensive supplementary meteorological data. The sensor measures path-integrated gas mass concentration (PIC by mass), which has units of kg/m^2; this can be imagined as collapsing all the gas along the path onto the area of the beam. It is more intuitively accessible to transform and quote these measurements as equivalent path-averaged concentrations by volume (PAC by volume), as this removes the effect of differing path lengths on the measurements and provides a number that reflects the average concentration along the path length, which can then be directly related to the average concentration (by volume) of the atmosphere as it is most commonly quoted. In the absence of any local CO_2 sources or sinks, the path-averaged concentrations of all the beams will be identically equal to the concentration of the naturally occurring background which is typically about 400 ppm during daytime.

In addition to concentration data, we continuously measure and record wind velocity and turbulence intensities (which drive the gas dispersion process) using a three axis ultrasonic anemometer (Figure 3 (c)). We also measure several other meteorological properties: temperature, pressure, solar insolation, humidity and precipitation using an automated weather station.

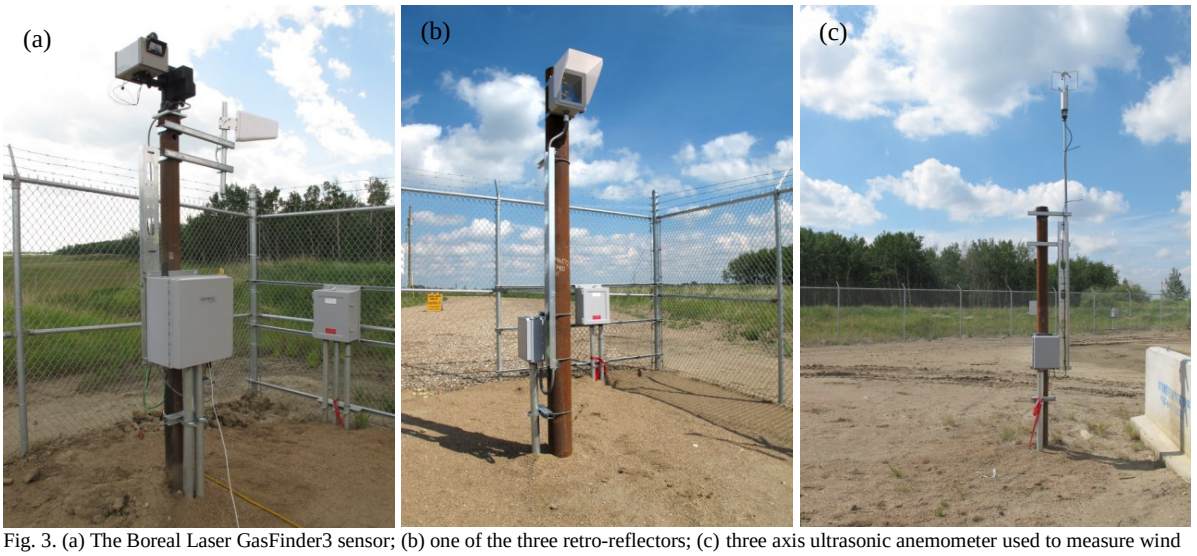

Fig. 3. (a) The Boreal Laser GasFinder3 sensor; (b) one of the three retro-reflectors; (c) three axis ultrasonic anemometer used to measure wind velocity and turbulence intensities.

2.3. Controlled CO_2 releases at Quest pad 8/19

We completed twenty-seven releases: twenty-four of these were 30 minute releases of ~300 kg/hr of CO_2 from one of four selected small area diffuse source locations; three other releases were of longer duration at lower release rates; two tests were set running overnight and included a fifth location. There were some short duration episodic emissions of gas during and after the first test related to balancing the thermal input to the pressure vessel. See Figure 4 for an overview of the experimental site and the controlled release locations. Release rates and exact locations are given in Table 1.

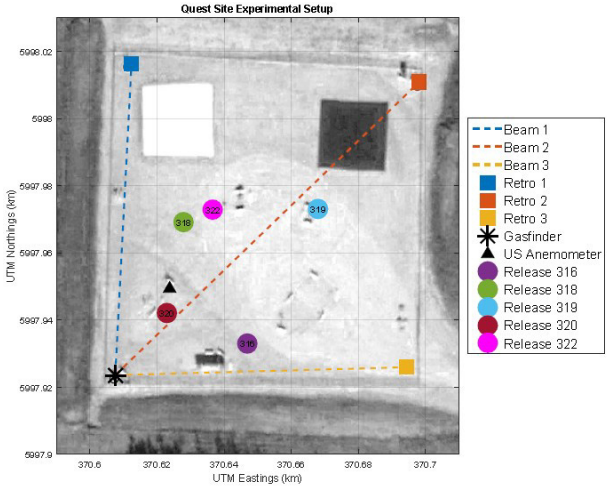

Fig. 4. Quest pad 8/19 experimental setup. Location of the Boreal GasFinder3 sensor shown with black star, squares indicate retro-reflectors and circles indicate controlled gas release locations.

2.3.1. Overview of experimental data

Our LightSource analysis method solves the inverse problem of where sources are and what emission rates are required to best explain the data. We use a Gaussian plume atmospheric eddy dispersion model [4] as our forward model; this provides an ensemble average of expected dispersion for steady state passive releases. The low concentration gas (hence no buoyancy effects) is passively transported by advection and diluted by turbulence in the atmosphere, which is a stochastic process. Effective dispersion prediction is fundamentally dependent on good measurements of: wind direction, wind speed and turbulence as well as concentration data. We require sufficient data to adequately reflect the steady state plume and hence be compatible with the steady-state ensemble average for concentration predicted by the dispersion model.

The data are grouped by release location, emission rate and duration of release, providing six data sets: 316, 316 (long), 318, 319, 320 and 322 (long). Each data set is compiled by concatenating all data sharing the same release location and release rate; data point color reflects beam under measurement for that period: beam1-blue, beam2-orange, and beam3-yellow (see Figure 5). The first 60 seconds data of each release are excluded to allow for equilibration to steady state of the plume crossing the pad from the release locations. The turbulence intensities reflect the variability of the wind direction over the averaging period, these values directly drive the dispersion process: as this is what mixes the gas with the air and dilutes it. All data for winds of less than 1m/s (for the averaging period) are excluded because of the disproportionately greater variability of low speed winds and weaker applicability of the dispersion model under these conditions. Wind data analysis showed that five of the six wind roses exhibit clear dominant wind directions: N-NW, S-SW and SE with a slight bias just discernible for N-NW and S-SW wind directions.

For two data sets we sustained releases for more than 30 minutes, in order to achieve this we had to reduce the mass emission rate; this is why two data sets are distinguished as being “long” (the reduced emission rates are given in Table 1). The other data sets comprise typically five or six periods of 30 minutes each, collected across the three day experimental program. The ordering of the data periods within a data set does not impact on the data analysis result.

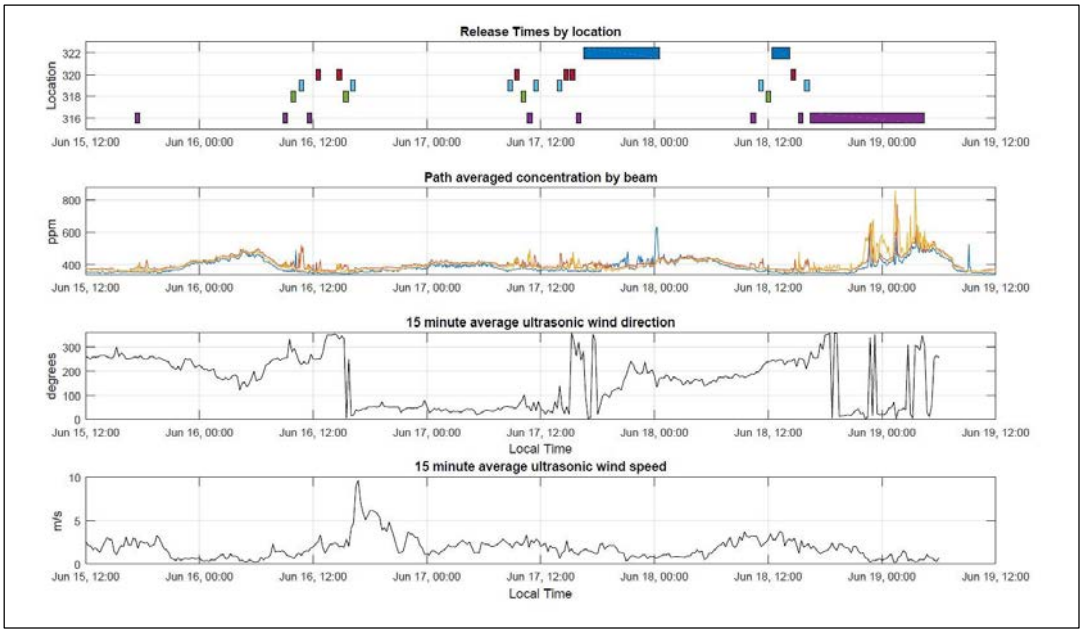

Fig. 5. Controlled release experiment: Top trace shows the time series of all controlled releases. The second trace plot shows path-averaged concentration, PAC, for Beam 1 (blue), Beam 2 (orange) and Beam 3 (yellow); concentration discrepancies imply an emission source. The sharp blue peak at the end is the CO_2 tank being vented at the end of the tests. The third trace shows wind direction and the bottom trace shows wind speed.

Table 1. Details of CO_2 releases from the experimental facilities at pad 8/19 at the Quest CCS site.

Date	Release number	Start time	Duration in hours	Release location	Latitude WGS-84	Longitude WGS-84	Flow meter drive pressure PSIG	Volume flow rate at NTP "air" SCF/min	Volume flow rate at NTP "CO2" SCF/min	Mass emission rate at NTP "CO2"
15/06/2015	1	17:12:30	0.50	316	54.113258°	-112.978860°	55	117	95.0	297.2
15/06/2015	venting	18:20:00	Intermittent	0						
16/06/2015	2	08:47:00	0.50	316	54.113258°	-112.978860°	56	117	95.0	297.2
16/06/2015	3	09:40:00	0.50	318	54.113577°	-112.979166°	56	117	95.0	297.2
16/06/2015	4	10:30:00	0.50	319	54.113623°	-112.978556°	55	117	95.0	297.2
16/06/2015	5	11:20:00	0.50	316	54.113258°	-112.978860°	55.5	118	95.8	299.8
16/06/2015	6	12:17:00	0.50	320	54.113333°	-112.979231°	55.5	120	97.4	304.9
16/06/2015	7	14:30:00	0.50	320	54.113333°	-112.979231°	56	119	96.6	302.3
16/06/2015	8	15:12:00	0.50	318	54.113577°	-112.979166°	56	119	96.6	302.3
16/06/2015	9	15:55:00	0.50	319	54.113623°	-112.978556°	56	119	96.6	302.3
17/06/2015	10	08:33:00	0.50	319	54.113623°	-112.978556°	55	120	97.4	304.9
17/06/2015	11	09:15:00	0.50	320	54.113333°	-112.979231°	55.5	118	95.8	299.8
17/06/2015	12	09:55:00	0.50	318	54.113577°	-112.979166°	55	118	95.8	299.8
17/06/2015	13	10:35:00	0.50	316	54.113258°	-112.978860°	55	118	95.8	299.8
17/06/2015	14	11:15:00	0.50	319	54.113623°	-112.978556°	55.5	118	95.8	299.8
17/06/2015	15	13:45:00	0.50	319	54.113623°	-112.978556°	56	118	95.8	299.8
17/06/2015	16	14:25:00	0.50	320	54.113333°	-112.979231°	56	118	95.8	299.8
17/06/2015	17	15:05:00	0.50	320	54.113333°	-112.979231°	56	118	95.8	299.8
17/06/2015	18	15:45:00	0.50	316	54.113258°	-112.978860°	56	116	94.2	294.7
17/06/2015	19	16:31:00	8.00	322	54.113613°	-112.979035°	40	90	73.1	228.7
18/06/2015	20	10:10:00	0.50	316	54.113258°	-112.978860°	55	116	94.2	294.7
18/06/2015	21	11:00:00	0.50	319	54.113623°	-112.978556°	54.5	116	94.2	294.7
18/06/2015	22	11:45:00	0.50	318	54.113577°	-112.979166°	55	116	94.2	294.7
18/06/2015	23	12:25:30	1.83	322	54.113613°	-112.979035°	29	110	55.1	172.5
18/06/2015	24	14:25:00	0.50	320	54.113333°	-112.979231°	55	116	94.2	294.7
18/06/2015	25	15:10:00	0.50	316	54.113258°	-112.978860°	55	116	94.2	294.7
18/06/2015	26	15:50:00	0.50	319	54.113623°	-112.978556°	54.5	116	94.2	294.7
18/06/2015	27	16:26:00	12.50	316	54.113258°	-112.978860°	30	113	57.3	179.2
19/06/2015	venting	08:40:00	intermittent	0						

We will present the results of our data analysis in the next section without going into the details of the inverse modeling which is beyond the scope of this paper and will be the subject of a future publication.

3. Results

Our analysis method expects emission sources to be at fixed locations and emit continuously at a constant rate. To avoid sustaining CO_2 emissions long enough for the wind to naturally vary over a substantial range of directions at one measured location, we have instead chosen several locations and performed several separate releases at each, so as to obtain sufficiently varied wind data at some locations to complete the analysis. Consequently, we need to

compile and analyze data for each release location separately. The resulting data set will therefore be much less constraining than that from a single sustained duration release, which would include a greater variety of wind directions, speeds and turbulence intensities as well as substantially more data points.

3.1. LightSource method

We illustrate the LightSource data analysis procedure using all the data collected from location 318. There is a balance of anomalous concentration data for several wind directions across different beams for this location; hence the problem is better constrained. There were four 30 minute release periods for location 318, each at 300 kg/hr; these were spread over the three days of experiments; so there are large time gaps in the data and step change discontinuities in the background concentration. These steps are clearly seen in Figure 6: where the black line of the top trace reflects the inferred shared ambient background concentration for the three beams. The code does not penalize sudden step changes in background concentration at the joins between different data sets, but does control it between consecutive measurements during the same release. Its setting is derived by long-term analysis of naturally varying background concentrations in the absence of emission sources. Hence the background concentration is held appropriately stable during each individual release period.

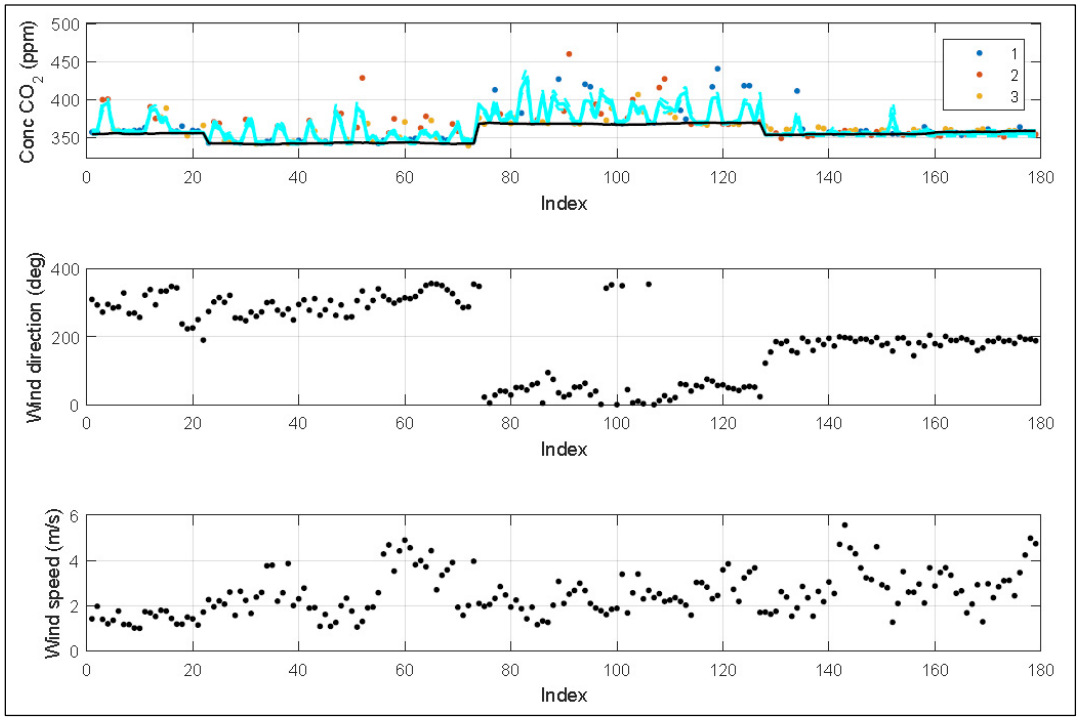

Fig. 6. The colored dots represent all the beam specific, 40 second averaged data for release location 318 that pass the selection criteria described in the text. The concentration data are path-averaged concentrations for the three beams. The black line is the inferred, background concentration shared by the three beams. The corresponding points on the two traces below are the wind direction and speed; each is associated with one concentration measurement, data are averaged over the same period. The cyan line is the model fit to the real concentration measurements.

The colored points of the top trace of Figure 6 show all the 40 second averaged beam data that pass the selection criteria, colors follow the previously established convention. The concentration data are path-averaged concentrations for the three beams, for the duration of all the sustained releases at location 318. About 15 minutes data from the first release period are excluded as they fail to satisfy the wind-speed threshold criterion. The continuous cyan line shows the model predicted concentrations for each measurement: i.e. the sum of the inferred

background concentration and the calculated concentrations resulting from the inferred source, for the dispersion conditions applicable to that beam measurement and associated wind and turbulence conditions at that measurement time. Those wind data are given in the second and third trace down in Figure 6 and are averaged over the same periods as the concentration data.

At first glance, the predicted concentrations do not appear a very good match to the measurements, but it should be noted that the dispersion model is a prediction of the ensemble average: the expectation averaged over a very large number of samples of what is essentially a stochastic process. So while individual results can look inaccurate, averaged over all the predictions made for all the locations considered and all the measurements encountered in the inversion, the average result is still adequate for our purposes. It is important that our dispersion model is in closed form - and hence simple- as it operates within our Markov Chain Monte Carlo (MCMC) scheme and the computational complexity must be kept manageable. It is also debatable just how much better more complex models perform in practice, given the underlying stochastic nature of atmospheric dispersion.

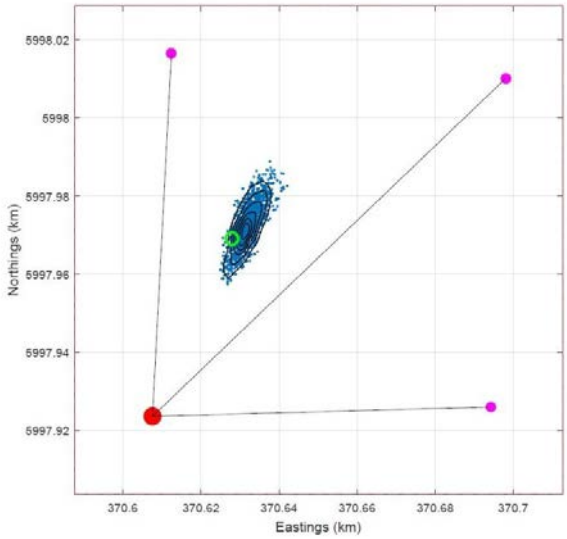

Fig. 7. LightSource result for the location of the emissions from release location 318. The real location is shown by the green circle. The real source was 2x2m (not shown to scale here); the blue patch represents the post burn in migration path of the 2000 post burn-in inferred source locations, overlain on this we give 2-dimensional kernel density plots to reveal the relative frequency of that location being selected: and hence an indication of relative merit of locations within the blue patch.

The LightSource estimates for the location of the source area 318 are given in Figure 7 shown as a blue patch; this is the path of the 2000 step post burn-in estimate of the source location generated by the MCMC process. Each of the 2000 points represents an equally justifiable individual estimate of the source location: but with different attributions of data-related uncertainty. In effect, uncertainties in wind speed, direction, turbulence intensity, inferred background concentration, inferred anomalous concentration and concentration measurements are mixed and matched to result in equally trustworthy individual results. Overlain on this blue patch are two dimensional kernel density contours. These reflect the relative number of the 2000 estimates falling within the respective contours which in turn provide insight into which locations are most frequently visited and hence to be preferred.

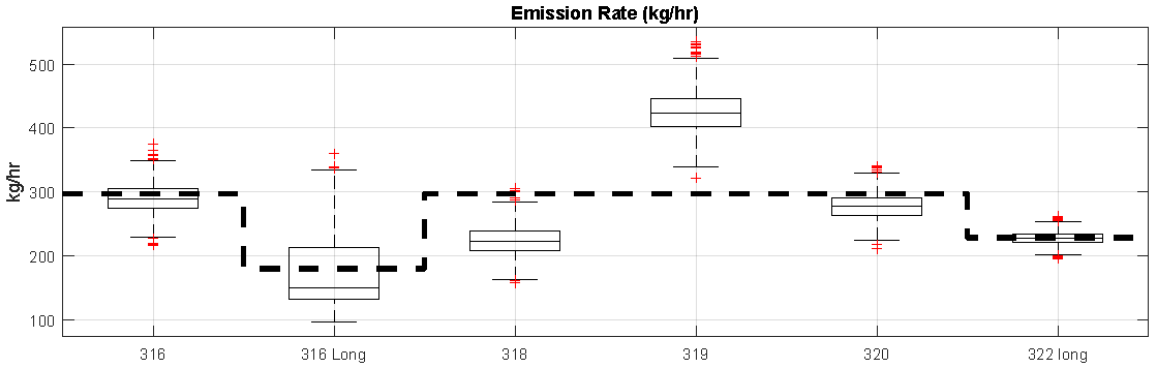

Fig. 8. Mass emission rate results for all source locations. The dashed line shows actual emission rates which were lower at long release times.

The inferred source emission rates for all locations are shown in Figure 8. The dashed line shows actual emission rates which were lower at long release times. This figure combines measurement related uncertainties and any systematically introduced errors, such as dispersion model inadequacies or systematic errors: such as wind speed/direction related errors.

The estimated source mass emission rates are broadly representative of the actual source emission rates for each of the data sets. However, the source location results are not as good as expected, based on previous field experiments with methane, even after allowing for the considerably greater difficulty of working with CO_2 (its 200 times greater background concentration and 10,000 times greater variability). Since the mass emission rate is determined by the range of the source from the where the concentration measurements are made, shortcomings in locating the sources will degrade the associated emission rates. The full inferred source location results reveal a progressive deterioration in location performance across the pad. This combined with our observations of plume behavior (via the bubble flow-visualization, and fence mounted simple wind direction indicators) prompted us to look more closely at our wind data.

We checked the alignment of both anemometers and found them to be good to within 1 or 2 degrees. When comparing the wind-speed data, however, we noticed that the Quest anemometer saw consistently greater wind-speeds and exhibited additional directional “lobes” that broadly correspond to the wind channels between the trees that border the Quest pad on three sides. The review revealed two deficiencies in our LightSource experimental set up: the ultrasonic anemometer was set up about a meter too high (exaggerating wind speeds) and some wind directions are strongly perturbed by the surrounding trees and hence poorly represented by measurements at our single anemometer.

Despite the limitations of these first experimental results with CO_2, there are several factors that will substantially improve LightSource performance in future installations:

- **New open-path gas sensor technology:** radically new open-path gas sensor systems will allow simultaneous and instantaneous measurements to be made along numerous beams. These sensors will not need to be steered to successive targets and dwell for minutes, as our current sensor does, but will continuously spin: allowing effectively simultaneous measurements on all beams. This will greatly improve our background concentration estimates and consequently, estimates of anomalous gas concentrations due to localized sources. These new sensors are also truly linear with better measurement precision and are immune to signal brightness fluctuations: meaning that longer beams will be possible and measurements will have both better measurement precision and immunity to scattering and atmospheric scintillation effects.
- **Beam geometry constraints relaxed:** it is the beam ends that provide the primary constraints for locating sources: as measured CO_2 plumes “fall off” the beam ends with changing wind direction, large changes in path-averaged concentration occur; indicating the center-line of the plume (i.e. the wind direction) is then aligned with the source and the beam end. Consequently, the more beam ends a system has the greater its

source reconstruction potential for a given measurement precision. The number of beam ends in our current system is limited by the time taken to align and suitably average a measurement along each beam. The relaxation of that constraint with the new sensor technology will make it feasible to include numerous beams and, additionally, to segment beams. This will dramatically increase the number of beam ends and thereby more precisely constrain the locations using what are effectively "range-resolved" beam sections.

- **Source reconstructions based on multiple LightSource installations at separate locations:** we are already simultaneously collecting and screening data for nine beams: three beams each on the three Quest pads. To date we have only reconstructed sources in the immediate vicinity of the pad where we have controlled release data, but as sensor precision increases we will be able to distinguish weaker and more remote sources and hence begin to triangulate persistent emission sources located between the pads using all the beam data. Ultimately we want to be able to detect the integrated total emissions from above the whole storage area.

4. Conclusions

We have demonstrated the first ever, automated, areal monitoring scheme for onshore Carbon Capture and Storage systems that is capable of automatically detecting the presence of CO_2 emissions from the well pads at the Quest CCS project. Our system has been able to estimate the locations and mass emission rates of the sources detected and these are in fair agreement with those of the actual sources.

The positive outcomes of these controlled CO_2 releases are that we have:

- Demonstrated that it is feasible to monitor an area for CO_2 emissions based on open-path optical path gas sensors and solution of the associated inverse gas dispersion problem
- Detected all twenty-seven controlled releases, as well as some weaker transient releases related to setting up and demobilizing the equipment
- Approximately located diffuse area source emission rates
- Provided useful estimates of the associated mass emission rates

Limitations to our current performance are understood and substantially better performance should be available from the current system with simple improvements to our experimental arrangements. The current performance is nonetheless superior to that available from any other system. Recent advances in open-path gas sensor technology offer dramatically enhanced measurement precision and acquisition speed. This is expected to result in greatly improved detection thresholds for emissions and matching performance improvements in locating and quantifying mass emission rates over larger areas.

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References

[1] Hirst B, Jonathan P, Gonzalez DelCueto F, Randell D, Kosut O. Locating and quantifying gas emission sources using remotely obtained concentration data. Atmospheric Environment, 74: 141-158, 2013.

[2] Levine ZH, Pintar AL, Dobler JT, Blume N, Braun M, Zaccheo TS, Pernini TG. The detection of carbon dioxide leaks using quasi-tomographic laser absorption spectroscopy measurement in variable wind. Published in Atmos. Meas. Tech., Copernicus Publications on behalf of the European Geoscience Union. 13 April 2016.

[3] Black TA, Whiticar MJ, Grant NJ, Nesic Z, Jassal RS, Jones H. Eddy-covariance measurements of carbon, water and energy fluxes at the Shell Quest site (pad 08-19); November 1, 2014-June30, 2015. Contract Report to Quest project, 2015.
[4] Draxler RR. Determination of atmospheric diffusion parameters. Atmospheric Environment, 10:99-105, 1976. ISSN 00046981.