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Rapid surface detection of CO₂ leaks from geologic sequestration sites

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Abstract

This study focuses on developing a method to characterize and detect leakage of carbon dioxide from a geologic sequestration site using a Picarro gas analyser, and to systematically evaluate the robustness of detection ability and optimize the data acquisition parameters by testing under varying conditions at the Zero Emissions Research and Technology field site in Bozeman, MT. It was determined (1) both ¹²CO₂ or ¹³CO₂ measurements provide equally good leak detection ability, (2) wind speed and direction does not limit detection ability with a sampling height less than 30 cm, and (3) δ¹³C measurements did not provide a reliable method for leak detection with our data acquisition strategy.

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1. Introduction

With carbon sequestration becoming a more viable carbon mitigation technology, new methods are needed to monitor and ensure such sites are functioning properly. There are several subsurface strategies being effectively employed [1], however, above surface detection of CO₂ is still a relatively new area of research. Subsurface

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monitoring techniques can effectively track plume migration but smaller leaks through highly permeable conduits such as faults, annular regions, or unsealed wells may go undetected. This study looks to develop and optimize an effective, robust, and cost efficient method for detecting below surface carbon dioxide leaks using rapid above surface monitoring of atmospheric concentrations of CO₂.

There are several reasons why above surface monitoring of CO₂ may be a beneficial addition to a comprehensive monitoring program. Firstly, safety around a site is paramount. While harmful occurrences have only been associated with volcanic events [2], there is still the possibility of localized cases where concentrations become high enough to be of concern [3]. Besides potentially harming animal and plant life, a leak would reduce the benefits of CCS on climate change mitigation. Finally, people must feel comfortable with carbon sequestration technologies before it becomes a viable solution for the global emissions problem [4]. Assuring that atmospheric concentrations stay normal during a sequestration project is one way of doing so.

Currently, there are several possible above surface monitoring techniques to compliment subsurface monitoring. These techniques include using eddy covariance towers, flux accumulation chambers, and atmospheric concentrations of CO₂. Eddy covariance technologies are used to measure gas flux in a localized area [5] and have been used with accuracy in different applications for some time now [6]. While accurate, this technique is limited in range and mobility [7]. Another detection method utilizes flux accumulation chambers. Like the eddy covariance measurements, these measurements are highly accurate but, depending on soil conditions, can vary greatly over 1 m [8]. Finally, atmospheric concentrations of CO₂ may be used to determine possible leak locations. One method, proposed by Krevor et al., used isotopic ratios of carbon dioxide to determine source attribution [9]. While anomalies in Krevor et al. have very large $\delta^{13}\text{C}$ isotopic signatures, this approach will not be successful if the isotopic signature of the source is similar to atmospheric concentrations.

This work expands on study by Krevor et al. which focused on developing a method to characterize and detect leakage of carbon dioxide from a geologic sequestration site based on a real-time detection system using a mobile sensing platform. The goal of this study is to systematically evaluate the robustness of detection ability and optimize the data acquisition parameters by testing under different conditions such as varying wind velocities, gas sampling inlet height, spatial resolution, sampling strategy, and isotope selection (e.g. ¹²C, ¹³C or $\delta^{13}\text{C}$).

This detection technique is designed to work in conjunction with other methods to ensure injected CO₂ stays underground. Ideally, this method would be used above possible subsurface plume locations determined from geophysical surveys and other subsurface techniques. If an anomaly is detected using this technique, more detailed above surface measurements from eddy covariance towers and fluxmeters could be made to quantify the leakage rate.

2. Methodology

The experimental design was divided into three distinct steps: collect data, identify anomalous atmospheric quantities, and relate anomalies in space to determine the most probable leak location.

2.1. Data collection

Data collection for this study was conducted at the Zero Emissions Research and Technology (ZERT) field site in Bozeman, MT, USA. The site is maintained by researchers at Montana State University (MSU) and was developed to study near surface transport and detection technologies through the controlled release of CO₂ [8]. The ZERT field site has a horizontal well approximately 1.8 m below the surface and has six separate packers in the center 70 m of the well (Figure 1). Flow rate from each of the packers can be controlled individually.

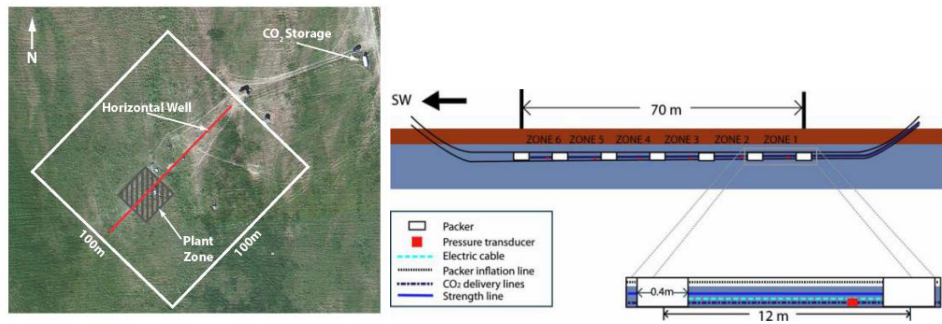


Fig. 1. ZERT field site map (left); Schematic of packers in horizontal well at the ZERT field site (right).

For this study, each of the packers released the same amount of CO₂ with a total leakage rate of 0.150 tons of CO₂ per day. This is equivalent to a 0.005 % leak from a 1 Mt per year CO₂ storage project. The injected CO₂ was sourced from a combustion process and had a ¹³C:¹²C ratio of 0.0104833:1 which corresponds to a δ¹³C of -67.1 ‰. By the time testing began field flux rates had reached steady state.

Topsoil at the ZERT field site consists largely of clays and silts with some sand that range between 0.2 and 1.2 m below the surface. Below the clay and silt layer there is a deposit of gravel that extends more than 5 m below the surface. Flora at the ZERT field site consists largely of alfalfa, clover, dandelions, thistle, and some other grasses. All of the plant life was cut below 10 cm except for a 20 m by 20 m section directly over the well that was deliberately uncut to study plant response due to leakage. There were several inaccessible areas of the site, the biggest of which was the area of uncut grass. No measurements were taken from this area to prevent damage to plant life. There were also two eddy covariance towers, one tri-level weather station, and a set of flux accumulation chambers that had to be avoided. The latter obstacles had relatively small footprints and were easily avoided.

Four types of data were collected as part of this study: CO₂ concentration data, location, wind velocity, and flux. All were used in concert to analyze the anomalies seen at the ZERT field site.

2.1.1. Concentration data

Carbon dioxide concentration data were taken using a Picarro G1101-i gas analyzer. The instrument was one of Picarro's first generation wavelength scanned cavity ring-down spectrometers. It was designed to provide concentrations of various greenhouse gases with high frequency, precision, and accuracy. This particular instrument was able to take ¹²CO₂ and ¹³CO₂ concentrations with a precision of 200 ppbv and 15 ppbv respectively. Concentration measurements were taken every one to three seconds.

2.1.2. Location data

Location data were taken every 1 s using a centimeter accurate Trimble GeoXH 6000 GPS unit. The unit was highly mobile and placed above the Picarro inlet hose for accurate concentration locations.

2.1.3. Wind data

Wind data were taken using a stationary Campbell Scientific Windsonic 1-L trilevel wind station near the center of the ZERT site. The instrument, managed by Montana State University, uses sonic anemometers to measure wind speeds in the horizontal direction. Wind measurements were simultaneously taken at three heights above the surface: 0.35 m, 0.85 m, and 1.5 m. Wind speeds were measured with a 0.01 m/s resolution and had less than a 2 % error. Wind directions were taken to 1 degree with an accuracy of ±3 degrees.

2.1.4. Flux data

Flux data at the ZERT field site were collected using a WEST Systems fluxmeter. Fluxes for 152 locations were taken within 30 m of the well. Measurements show several locations of higher flux, or hot spots, located along the well (Figure 2). These hotspots are thought to be caused by CO₂ migrating to the highest point of each packer and then moving vertically upward to the surface. Flux measurements were used to validate the locations of possible anomalies detected with the concentration data.

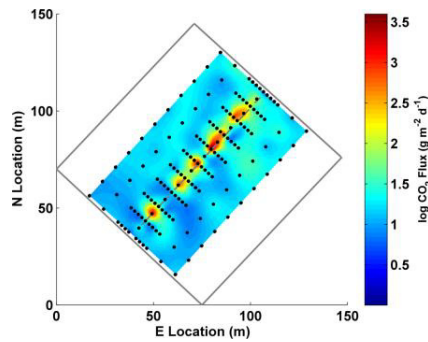


Fig. 2. Log soil CO₂ flux map at the ZERT field site under 0.15 t/day leakage with black points representing locations at which measurements were taken using a flux accumulation chamber

2.1.5. Data collection methods

All the concentration data reported here were collected using mobile sampling. During mobile testing, the Picarro instrument, GPS unit, and power source were mounted to a typical garden wagon. The wagon was manually pulled around the ZERT field using varying transects with speeds between 0.5 and 2 m/s. Transit velocities were calculated using the GPS data.

2.2. Detecting anomalous atmospheric quantities

The second step in determining possible leak locations is detecting anomalous atmospheric quantities as a function of time. Several quantities were examined, including ¹²CO₂, ¹³CO₂, and δ¹³C. Two methods, using static and adaptive backgrounds were used to determine possible concentration anomalies (Figure 3). Both methods determine anomalies based on concentration above background. The two methods differ, however, in determination of the background. The first method uses a static background, while the second method uses an adaptive background. The static background is determined by typical concentrations seen away from the well and stays constant, or static, throughout the survey. This is in contrast to the adaptive background method which is determined by taking the median concentration of temporally neighboring measurements and will tend to change throughout each survey. In this study, the adaptive background concentration was calculated using 21 points: 10 points in the future, 10 in the past, and the current location. The length it takes to collect 21 measurements is approximately the length of spatial background variations at the ZERT field site (50 m).

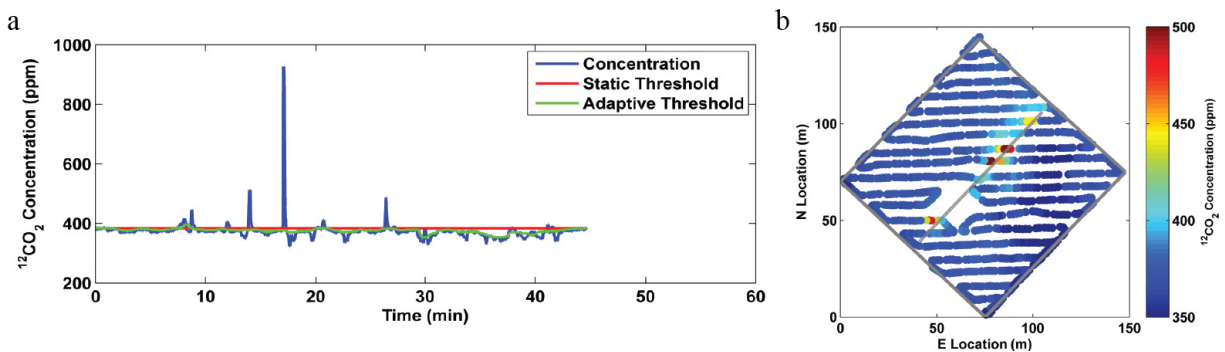


Fig. 3. Example ¹²CO₂ dataset from August 19, 2013; (a) concentration as a function of time with static and adaptive thresholds shown for reference; (b) concentration map.

A threshold above the established background concentration is then determined. Anything above this threshold was considered an anomaly while anything below was considered background. In both cases, the threshold above

background was based on natural concentration variability seen at the ZERT field site. We selected two standard deviations above background concentrations as the threshold for identifying an anomaly. The static threshold was 383.3370 ppm and 4.3416 ppm for $^{12}\text{CO}_2$ and $^{13}\text{CO}_2$ respectively while the adaptive threshold constantly changed.

Finally, anomalous $\delta^{13}\text{C}$ values were determined using thresholds similar to the static and adaptive thresholds used for concentrations. Typical atmospheric and soil $\delta^{13}\text{C}$ values are approximately -8 ‰ and -27 ‰ respectively [9]. Any $\delta^{13}\text{C}$ value below two standard deviations from the background is considered anomalous.

2.3. Locating concentration anomalies

The last step in determining possible leakage location is spatially relating anomalies detected in the previous step. At this point, each $^{12}\text{CO}_2$, $^{13}\text{CO}_2$, and $\delta^{13}\text{C}$ value is either anomalous or within the range of background values (Figure 4a-b). The irregularly gridded anomaly data are then rasterized using a linear interpolation to a grid with 0.5 m by 0.5 m cells. An example of this is provided in Figure 4c-d. Each cell was rounded to indicate its likely background or anomalous state. Linear interpolation was chosen over other types of spatial interpolation as it requires no manual input which is essential in an autonomous system.

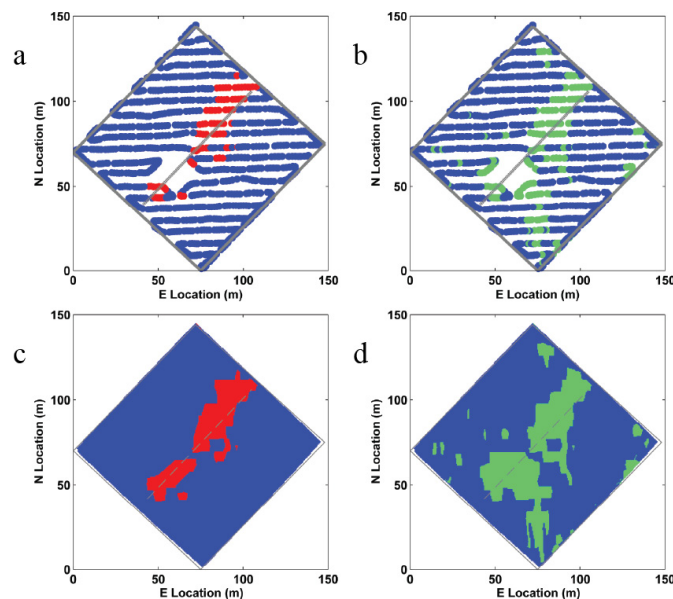


Fig. 4. Example dataset from August 19, 2013 using $^{12}\text{CO}_2$ to determine anomalies; (a) static threshold anomalies in space; (b) adaptive threshold anomalies in space; (c) rasterized anomalies from static threshold test; (d) rasterized anomalies from adaptive threshold test.

3. Results

3.1. Leak detection ability

The ability to detect and locate the leak was determined using 18 mobile surveys that covered various sections of the ZERT site. They were conducted at different times of day under varying wind and weather conditions. Additionally, the mobile surveys were conducted at inlet heights of 3 and 30 cm using various sampling patterns and speeds (0.75-1.00 m/s). The areas covered in these surveys can be seen in Figure 5.

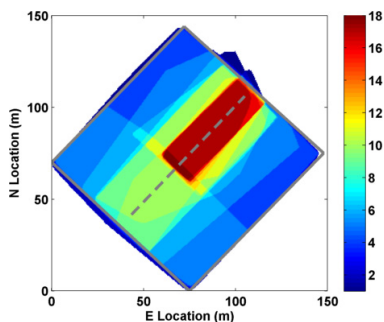


Fig. 5. Number of surveys taken in each area of the ZERT field site.

Consistent leak detection was seen around hotspots using $^{12}\text{CO}_2$ and $^{13}\text{CO}_2$ concentrations. Figures 6 and 7 show the percentage of times an anomaly was detected in each area. This was calculated by dividing the number of positive detections by the number of surveys conducted in that area and was done for $^{12}\text{CO}_2$, $^{13}\text{CO}_2$, and $\delta^{13}\text{C}$ using both static and adaptive thresholds.

The static threshold method using either $^{12}\text{CO}_2$ or $^{13}\text{CO}_2$ showed very consistent leakage detection around the source. Detection within 5 m of a hotspot was near 100% while detection 7.5 m downwind was greater than 60%. Even under varying conditions, leak detection is almost certain due to a high detection percentage near the well. There are also some detected leaks farther than 10 m from the well which are thought to be caused by plumes downwind of the source or false detections. The most likely cause for false detections using the static threshold method are temporally variable background concentrations. This phenomena can be seen during dawn and dusk in which photosynthesis begins to effect background CO_2 concentrations. $\delta^{13}\text{C}$ measurements were too variable (2 standard deviations = 18.16 per mil) to provide consistent detection using the static threshold method. The best possible detection was around 40% near the largest hotspot. This can be explained by the asynchronous measurements of $^{12}\text{CO}_2$ and $^{13}\text{CO}_2$. When concentrations are changing quickly, which they do under these sampling conditions, a large amount of error is introduced into the calculated value for $\delta^{13}\text{C}$.

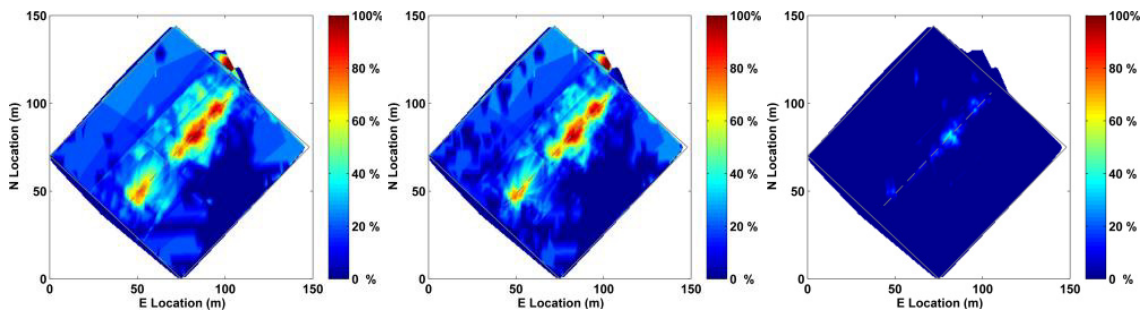


Fig. 6. Detection percentage using static threshold method for $^{12}\text{CO}_2$ (left), $^{13}\text{CO}_2$ (center), and $\delta^{13}\text{C}$ (right).

The adaptive threshold method offered similar results to the static threshold method. Detection was near 100% within 5 m of the well and greater than 60% 7.5 m downwind of the source. Again, there were small areas of detection farther from the well either due to downwind plumes or false detection. Any false detections using the adaptive threshold were thought to be caused by spatially variable background concentrations caused by transitions between vegetation types. Like the static threshold method, $\delta^{13}\text{C}$ measurements provided unreliable detection at best.

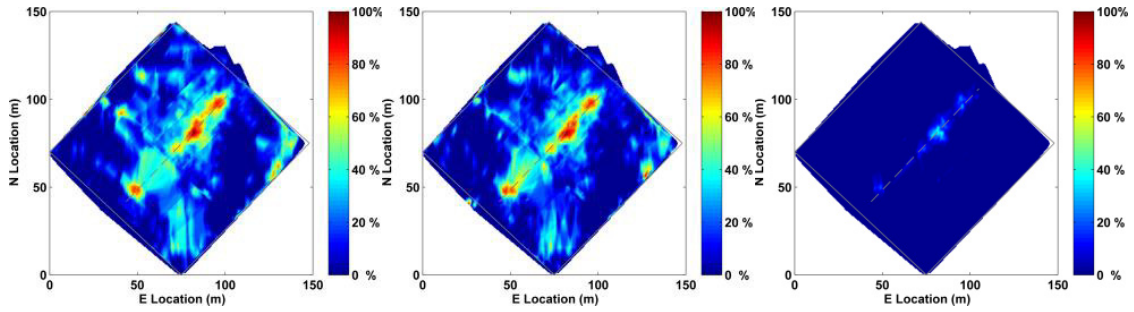


Fig. 7. Detection percentage using adaptive threshold method for $^{12}\text{CO}_2$ (left), $^{13}\text{CO}_2$ (center), and $\delta^{13}\text{C}$ (right).

3.2. Effect of wind and optimal sampling height

Winds during mobile testing were variable and ranged between 0 and 2.5 m/s in all directions. As shown from the high probability that leaks are correctly detected and located in Figures 6-7, it can be inferred that varying wind conditions with speeds up to 2.5 m/s do not interfere with detection ability. The subsequent analysis is carried out by comparing the effect of wind speed on leak detection with gas sampling heights. Both used the static detection criteria for comparability. The surveys were taken at a sampling height of 0.30 m and 0.03 m in high and low wind conditions. While the survey taken at 0.30 m seen in Figure 8 had a larger footprint under low wind conditions, there was a 66% reduction in footprint area under higher winds. The survey taken at 0.03 m seen in Figure 9 had a smaller footprint under lower wind conditions but only showed a 5.7% reduction in footprint size under windier conditions. This suggests that a lower sampling height will provide more consistent detection under various wind conditions.

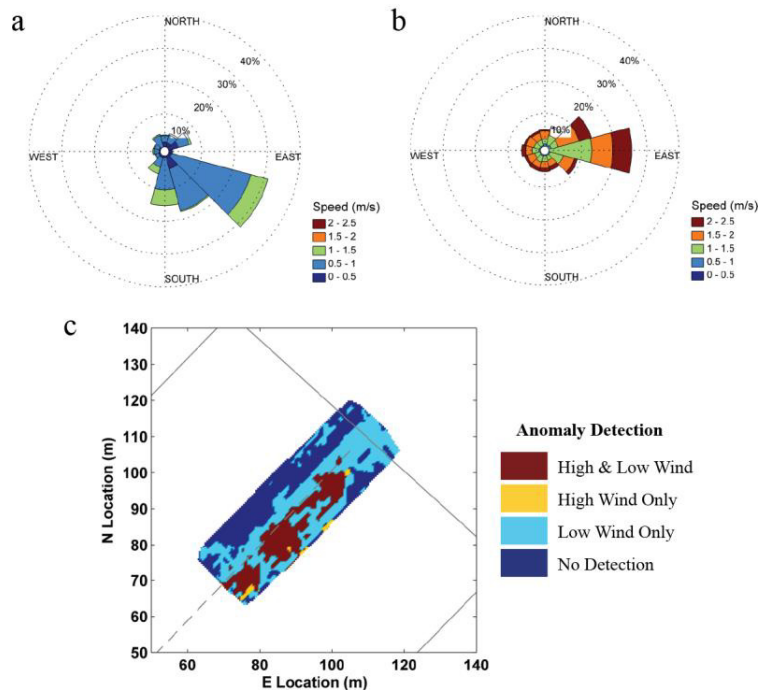


Fig. 8. Wind rose for survey at 0.30 m for low wind (a) and high wind (b) conditions; (c) Comparison of anomaly detection at 30 cm under low and high wind conditions.

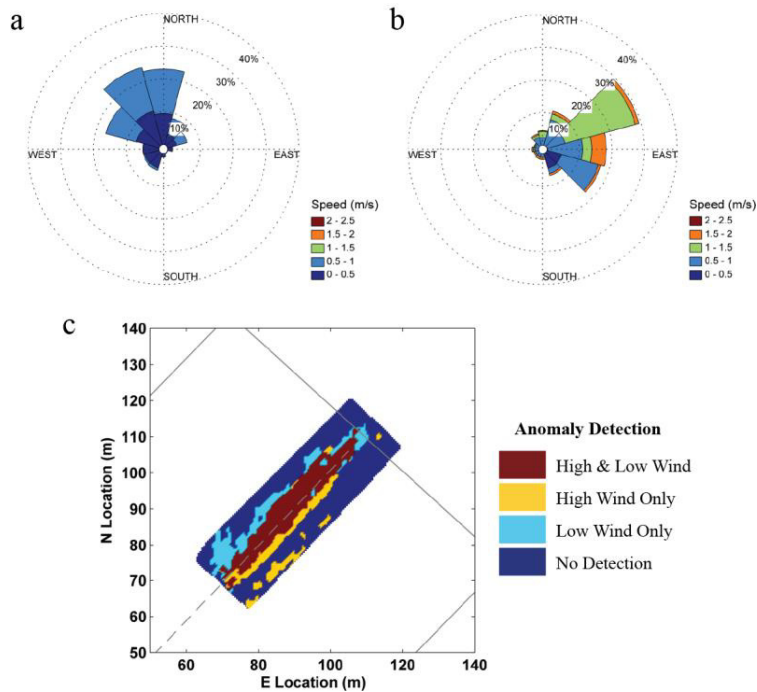


Fig. 9. Wind rose for survey at 0.03 m for low wind (a) and high wind (b) conditions; (c) Comparison of anomaly detection at 3 cm under low and high wind conditions.

3.3. Instrument velocity

The influence of sample acquisition was also tested. Several smaller surveys bisecting the ZERT site perpendicular to the well were taken. The surveys were taken at 0.50, 1.00, 1.25, and 2.00 m/s and passed over the source twice. Each survey detected anomalies over the source. This suggests an anomaly will be seen if the distance travelled between readings is less than the size of the plume.

4. Conclusion

The objective of this research was to (1) quantify the probability that CO₂ leaks can be detected and located using a real time detection system using an above ground, mobile sensing platform and (2) optimize data acquisition parameters and data analysis methods for application to real-world settings. The method was designed to be rapidly deployable, efficient, cost effective, and would fill the monitoring gap between subsurface and stationary above ground measurements. In addition, the effects of wind, sampling height, and instrument velocity on detection ability were investigated to ensure accurate and efficient detection. There are several conclusions about the using a mobile high-precision gas analyzer that can be drawn from the results.

- Leak Detection: Surveys sampling ¹²CO₂ or ¹³CO₂, around a 0.15 t/day subsurface leak of carbon dioxide can be detected with near 100% certainty if within 5 m of a leak. Surveys using δ¹³C were not able to consistently detect leak locations.
- Effect of Wind: Wind speeds up to 2.5 m/s do not have a large effect on leak detection ability. Wind speeds greater than 2.5 m/s could not be tested as no speeds above 2.5 m/s were experienced during testing
- Optimal Sampling Height: The optimal sampling height is closest to the ground as possible. Sampling near the surface provides the most consistent detection ability under varying wind conditions, however, under no/light wind conditions a sampling height of 0.30 m was shown to increase the detection area.

- Instrument Velocity: Instrument velocity does not have an effect on detection ability using $^{12}\text{CO}_2$ and $^{13}\text{CO}_2$ detection criteria unless the instrument travels the length of an anomaly between measurements.
- Mobile, high precision measurements of the stable isotopes ^{12}C and ^{13}C provide an excellent method for quickly and efficiently detecting and locating CO_2 leaks and have great potential for monitoring geological storage sites.

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