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Authors
Lewicki, J.L.
Oldenburg, C.
Dobeck, L.
et al.

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Surface CO$_2$ leakage during the first shallow subsurface CO$_2$ release experiment

J.L. Lewicki$^1$, C.M. Oldenburg$^1$, L. Dobeck$^2$, and L. Spangler$^2$

$^1$ Earth Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., MS 90-1116, Berkeley, CA 94720 USA, jllewicki@lbl.gov

$^2$Department of Chemistry and Biochemistry, Montana State University, 108 Gaines Hall, PO Box 173400, Bozeman, MT, 59717 USA
Abstract. A new field facility was used to study CO$_2$ migration processes and test techniques to detect and quantify potential CO$_2$ leakage from geologic storage sites. For 10 days starting 9 July 2007, and for seven days starting 5 August 2007, 0.1 and 0.3 t CO$_2$ d$^{-1}$, respectively, were released from a ~100-m long, sub-water table (~2.5-m depth) horizontal well. The spatio-temporal evolution of leakage was mapped through repeated grid measurements of soil CO$_2$ flux ($F_{CO_2}$). The surface leakage onset, approach to steady state, and post-release decline matched model predictions closely. Modeling suggested that minimal CO$_2$ was taken up by groundwater through dissolution, and CO$_2$ spread out on top of the water table. $F_{CO_2}$ spatial patterns were related to well design and soil physical properties. Estimates of total CO$_2$ discharge along with soil respiration and leakage discharge highlight the influence of background CO$_2$ flux variations on detection of CO$_2$ leakage signals.

1. Introduction

As geologic carbon sequestration gains momentum as a viable strategy to mitigate climate change associated with elevated CO$_2$ concentrations in the atmosphere, the number of large, industrial-scale and smaller-scale pilot CO$_2$ injection projects has increased [e.g., International Energy Agency, 1997, 2004; IPCC, 2005]. While the purpose of geologic carbon sequestration is to trap CO$_2$ underground, CO$_2$ has the potential to leak from the storage site along permeable pathways such as well bores or faults to the near-surface environment. The technical community must therefore demonstrate the ability to detect, characterize, mitigate, and remediate CO$_2$ leakage from geologic CO$_2$ storage sites to
satisfy public concerns about safety and environmental impact of geologic CO\textsubscript{2} storage. In particular, near-surface detection of CO\textsubscript{2} leakage could be challenging due to the large variation in natural background CO\textsubscript{2} fluxes arising from biological processes [e.g., Lewicki \textit{et al.}, 2005]. A new facility was recently built in an agricultural field at Montana State University by the Zero Emissions Research and Technology (ZERT) Project to release CO\textsubscript{2} into the shallow subsurface from point and line sources that emulate leakage along, e.g., abandoned wells or faults. This is to our knowledge the first facility that provides the opportunity to study CO\textsubscript{2} migration processes and to test techniques to detect and quantify potential CO\textsubscript{2} leakage from geologic storage sites.

In July and August 2007, two controlled releases of CO\textsubscript{2} were carried out at different rates from a shallow horizontal well. Changing meteorological conditions and associated soil microclimate and plant phenology over this timeframe led to varying levels of background biological fluxes within which leakage signals evolved. We conducted numerical modeling of the CO\textsubscript{2} releases to elucidate CO\textsubscript{2} migration processes and predict the magnitude and geometry of CO\textsubscript{2} leakage signals. We then carried out detailed measurements of soil CO\textsubscript{2} flux ($F_{\text{CO}_2}$) along a grid at varying distances from the well to characterize the spatio-temporal evolution of both CO\textsubscript{2} leakage and background biological (soil respiration) fluxes, and to quantify surface CO\textsubscript{2} leakage rates. Here, we (1) present and compare field measurement and modeling results of what is to our knowledge the first-ever CO\textsubscript{2} shallow-release experiments aimed at studying surface leakage from geologic storage projects, and (2) discuss implications of the results for detection of surface leakage.
2. Field Site and Experimental Design

The CO$_2$ release experiments were conducted at Montana State University, at the Montana Agricultural Experiment Research Center in Bozeman, MT. The study site was a \( \sim 0.12 \) km$^2$ nearly flat field, with prairie grasses, alfalfa, and Canadian thistle. Here, a \( \sim 30 \) cm-thick clay topsoil overlies a \( \sim 20 \) cm-thick clayey silt layer, which overlies an alluvial sandy cobble with 10-25 cm diameter cobbles. A N45E-trending horizontal well with a 73-m long central slotted (perforated) section and 15- and 12-m long unslotted sections on the sloping NE and SW ends, respectively, was installed in the field. The slotted section was located at \( \sim 1.3-2.5 \) m depth within the alluvial sandy cobble and was divided into six \( \sim 12 \) m long zones separated by 0.4-m wide inflatable packers (Figure 1a). The water table depth was \( \sim 1.6 \) m, resulting in sub-water table CO$_2$ releases. From 9-19 July 2007 (Release 1), and from 5-10 August 2007 (Release 2), 0.1 t CO$_2$ d$^{-1}$ and 0.3 t CO$_2$ d$^{-1}$, respectively, were released from the well evenly from each of the six slotted zones. The 0.1 t d$^{-1}$ rate was chosen based on numerical simulations to provide a challenging detection problem while still ensuring that injected CO$_2$ would reach the ground surface. The 0.3 t d$^{-1}$ rate was chosen to obtain a larger surface flux for demonstration purposes.

2. Methods
The simulator TOUGH2/EOS7CA [Pruess et al., 1999; Oldenburg and Unger, 2003; 2004] for modeling subsurface migration of water, CO$_2$, and air is used to model CO$_2$ releases into the shallow subsurface. Properties of the two-dimensional (2D) model oriented transverse to the horizontal well are shown in Table 1. In all cases, the initial condition is a gravity-capillary steady state with zero rainfall infiltration, constant pressure at the top and bottom, and no groundwater flow. Note the larger moisture retention capacity of the soil leads to an initial condition with a capillary barrier at the soil-cobble interface. A shallow vertical-well CO$_2$ injection test was conducted at the field site in October 2006 to observe injectivity and surface CO$_2$ flux of the soil-cobble system. Accumulation chamber measurements of F$_{CO_2}$ for this test were used as constraints to fit model permeabilities to the two-layer soil-cobble system. The high calibrated permeability of the soil (Table 1) likely arises from cracks and root casts that create macropores through which soil gas and atmospheric air readily flow. Fitted soil and cobble properties were then used in forward models of the two horizontal well releases.

F$_{CO_2}$ was measured using a WEST Systems Fluxmeter (WEST Systems, Pisa, Italy) based on the accumulation chamber method [Chiodini et al., 1998], with accuracy and repeatability of $-12.5\%$ [Evans et al., 2001] and $\pm10\%$ [Chiodini et al., 1998], respectively. F$_{CO_2}$ was measured at 1-m spacing along the surface well trace on 17-18 July, and 7-8 August 2007 (Figure 1), and repeatedly on a daily basis at 2.5 to 10-m spacing on grids from 7-16 July and from 9-12 August 2007 (Figure 2). F$_{CO_2}$ measurements were made between 03:00 and 14:00 on any given day. F$_{CO_2}$ maps were
interpolated from grid measurements using a minimum curvature spline technique. While this method produces a relatively smooth surface, it remains faithful to the original sample data. Geostatistical interpolation/simulation methods [e.g., Lewicki et al., 2005] were inappropriate for our $F_{\text{CO}_2}$ datasets due to poor spatial correlation on the grid measurement scale and lack of stationarity. Total CO$_2$ discharge ($D_{\text{tot}}$) was estimated for each grid dataset by calculating the declustered mean $F_{\text{CO}_2}$ using GSLIB [Deutsch and Journel, 1998] and multiplying it by the total measurement area (7700 m$^2$).

3. Results

For Release 1, numerical simulations predicted surface breakthrough of CO$_2$ leakage after 1.5 days (Figure 3). Modeled leakage flux at the surface above the well then reached near-steady state on ~Day 6 of Release 1; however, flux continued to increase very gradually over the remainder of the release period. Simulated leakage flux declined sharply by ~50% over the first day following the end of Release 1, and then declined more gradually to low (near-zero) values by the beginning of Release 2. For Release 2, surface breakthrough was predicted to occur more quickly, and leakage flux above the well was predicted to reach steady state after only ~3 days. The decline in simulated leakage flux was sharp (by >90%) over the first day following the end of Release 2, and then more gradual over subsequent days.

Cross-sections of simulated subsurface CO$_2$ concentrations and corresponding cross-well profiles of surface CO$_2$ flux are shown in Supplement 1 for Day 8 of Releases 1 and 2.
(i.e., near-steady state conditions). On Day 8 of both releases, mushroom-shaped subsurface CO₂ plumes were predicted (Supplement 1b and c), with CO₂ spreading along the top of the water table, and maximum concentrations of >0.9 mass fraction CO₂ within the cores of the plumes. Profiles of predicted surface CO₂ flux were symmetrical around the surface well trace (Supplement 1a and b) and, if extrapolated along the length of the well, would result in constant longitudinal leakage flux. The predicted width of the subsurface CO₂ plume was greater for Release 2 than 1 (Supplement 1b and d), which resulted in a wider zone of surface leakage fluxes (i.e., spreading to ~5 m from the well trace, versus to 2.5 m) (Supplement 1a and c). Maximum surface leakage fluxes simulated for Releases 1 and 2 were ~400 and 1200 g m⁻² d⁻¹, respectively. We emphasize that the simulations were predictive, i.e., carried out before the horizontal shallow-release experiments, with permeability calibration based on the earlier vertical well injection test.

Figure 2 shows the spatio-temporal evolution of F_CO₂ measured during the timeframes of Releases 1 and 2 and Figure 3 shows the corresponding CO₂ discharges. There was no evidence of F_CO₂ related to leakage at distances >7.5 m from the well trace. Consequently, to estimate background (soil respiration) CO₂ discharge (D_back) for each grid dataset, we calculated the mean F_CO₂ for distances 10-30 m from the well trace, and assuming this F_CO₂ was representative of background F_CO₂ for the entire grid area, multiplied it by 7700 m². The CO₂ discharge associated with leakage from along the well (D_leak) was then estimated as D_tot – D_back (Figure 3). A decrease in background F_CO₂ was observed over the two days preceding Release 1, which continued during the first day of
Release 1 when no evidence of leakage was observed at the surface (7-9 July 2007); Figures 2a-c and 3). Breakthrough of CO₂ at the surface, indicated by elevated FCO₂, was observed at a single point along the well trace on Day 2 of Release 1 and remained relatively stable to Day 3 (Figure 2d and e). On these days, Dtot remained similar to that observed on Day 1 of the release, while Dback decreased, and Dleak increased (Figure 3). Then, elevated FCO₂ was measured at six point sources aligned along the well trace on Day 4 of the release (Figure 2f). The position of these leaks remained stable over the next six days, while the magnitude of FCO₂ increased from Day 4 to 6, to remain relatively constant until Day 10 (Figures 2g-j and 1b). Maximum FCO₂ was ~1600 g m⁻² d⁻¹. From Day 4 to 8, Dtot was highly variable and did not exceed values measured prior to Release 1. Changes in Dtot over this time period generally followed changes in Dback, while Dleak increased to remain relatively stable at ~0.1 t d⁻¹ from day 6 to 8 (Figure 3). Figure 1a and b illustrate the spatial relationship of the FCO₂ leakage anomalies to well design. Five of the six FCO₂ peaks measured along the well trace were located above the well packers (packers 6, 4, 3, 2, and 0) and tended to be located above the higher elevation end of the slotted well sections. An exception to this pattern is the FCO₂ peak measured above the unslotted section on the far NE end of the well.

FCO₂ measurements began on Day 5 of Release 2 and showed similar surface leakage patterns as those observed during Release 1 (Figures 1c and 2k and l). However, the magnitude of FCO₂ measured along the well trace was higher (maximum = 6000 g m⁻² d⁻¹) and a greater degree of spreading of leaking CO₂ was observed both along and away from the well trace relative to Release 1. Dtot and Dleak were ~0.45 and 0.33 t d⁻¹, respectively,
on Days 7 and 8 of Release 2, while $D_{\text{back}}$ remained relatively low. $F_{\text{CO2}}$, $D_{\text{tot}}$, and $D_{\text{leak}}$ showed large declines on Day 1 following the end of Release 2 and dropped to near-background values on the second day after the release (Figures 2m and n and 3).

4. Discussion and Conclusions

We present $F_{\text{CO2}}$ measurements and numerical simulations associated with the first-ever CO$_2$ release experiments from a subsurface line source. Model predictions of the evolution of the surface flux leakage signals were closely matched by field measurements of $F_{\text{CO2}}$. For example, surface breakthrough of CO$_2$ was predicted to occur 1.5 days after the start of Release 1, and was observed on Day 2 (precise breakthrough time was not recorded by daily grid measurements). A rapid drop of the surface leakage signal was both predicted and observed following the end of Release 2 (Figures 2d, m, n and 3). Also, assuming that the temporal evolution of leakage CO$_2$ flux over the well and $D_{\text{leak}}$ should be similar, both predicted and observed leakage signals reached near-steady state on Day 6 of Release 1 (Figure 3). Finally, the observed extent of CO$_2$ spreading away from the well (i.e., a maximum of 2.5 to 5 m for Release 1 and 5 to 7.5 m for Release 2) at near-steady state conditions was close to that predicted by models (Figures 2j and l and 4a and c).

As suggested by numerical models, while some CO$_2$ spreading likely occurred on top of the water table, little CO$_2$ was lost to (dissolved in) the groundwater system during the releases (Figures 4b and d). As a result, the groundwater system minimally attenuated
CO₂ flow to the surface, D_{leak} values on Days 6-8 of Release 1 and Days 7-8 of Release 2 were close to CO₂ release rates (Figure 3), and CO₂ spreading away from the well was limited. Also, the relatively fast predicted and observed breakthrough time of CO₂ to the surface during Release 1 and decline of F_{CO₂} to near-background values following the end of Release 2 were likely due in part to high soil permeability caused by macropores allowing for rapid exchange of soil and atmospheric gases.

There were key differences between predicted and observed leakage flux signals. First, numerical simulations were oriented transverse to the well and therefore did not model the observed multiple point-source leakage signals aligned along the well trace, which showed some connection to one another on Days 7 and 8 of Release 2 (Figures 1 and 2). Second, the maximum predicted leakage fluxes above the well were lower than those measured during Releases 1 and 2 due to the longitudinal averaging implicit in the 2D transverse model. The spatial distribution of observed leakage fluxes was strongly correlated with the well design (Figure 1). CO₂ likely flowed from relatively low to high elevation within the well injection zones until it encountered the barriers of packers 6, 4, 3, 2, and 0. It probably then flowed upward to the surface, leading to concentrated areas of relatively high-magnitude surface leakage. Unmapped zones of high soil permeability may have further focused CO₂ flow. The far NE F_{CO₂} peak measured above the unslotted well section was likely due to CO₂ flow to the surface along the outside of the well bore, an unexpected process not included in the numerical model. Higher vertical pressure gradients were probably established by the higher CO₂ release rate of Release 2, leading to more direct flow of CO₂ from its release points to the surface and a more
longitudinally continuous surface leakage signal, relative to Release 1. While the intent of the release experiments was to create a longitudinally uniform leakage pattern, the effects of well design and soil physical properties likely created signals more realistic of leakage along partially-sealed faults or fractures, where fluids migrate through discrete pathways to the surface. Leakage along such features may actually be more likely at sites selected for CO2 storage, where, if present, faults will probably be inactive and largely sealed.

The grid used for chamber measurements included measurement points close to and away from the horizontal well, allowing us to quantify CO2 emissions from background soil respiration processes separately from leakage. We observed relatively high $D_{\text{back}}$ on 7 July 2007, followed by a decrease at about the same rate as the increase in $D_{\text{leak}}$ (Figure 3). Consequently, $D_{\text{tot}}$ was variable during Release 1, but did not exceed values measured prior to the release. A rainstorm occurred on the evening of 6 July 2007, during otherwise dry and hot (average daytime temperature = 22°C) weather conditions. The decrease in $D_{\text{back}}$ following the rainstorm was likely due to a decline in soil moisture content and associated plant and microbial activity. A primary challenge of near-surface detection of potential CO2 leakage from geologic storage sites is to discern a leakage signal within background CO2 variability. This could be difficult if the signal is of very small magnitude and/or spatial extent [e.g., Lewicki et al., 2005]. Both Releases 1 and 2 resulted in high-magnitude leakage relative to background CO2 fluxes, but the overall areas of the anomalies were small relative to the grid area. As a result, when background $F_{\text{CO2}}$ is high (e.g., during the growing season, or after rain events during dry periods), it
can mask leakage \(F_{\text{CO}_2}\). This effect was clear during Release 1, when considering \(D_{\text{tot}}\), and would be stronger if one were attempting to detect leakage signals within a larger, reservoir-scale area. Since background \(F_{\text{CO}_2}\) was relatively low during Release 2, \(D_{\text{tot}}\) was clearly discernable from \(D_{\text{back}}\) measured prior to Release 1. Because the point-measurement nature of the chamber method allows mapping of the spatial distribution of \(F_{\text{CO}_2}\) and we measured \(F_{\text{CO}_2}\) on a spatial scale fine enough to capture the leakage signal, leakage was visible in \(F_{\text{CO}_2}\) maps during both Releases 1 and 2 (Figure 2). Use of a CO\(_2\) measurement technique, however, that averages over a relatively large area (e.g., eddy covariance) would likely have rendered CO\(_2\) leakage detection during Release 1 difficult.

Our results emphasize the importance of (1) careful characterization of background CO\(_2\) variability prior to CO\(_2\) injection into the storage reservoir, (2) limitation of the total area of investigation by focus on features most susceptible to leakage (e.g., wells, faults), and (3) use of a variety of complementary CO\(_2\) measurement techniques in a program of storage site monitoring. Overall, the new ZERT CO\(_2\) release facility provides an excellent opportunity to study CO\(_2\) migration processes in the near-surface environment and develop integrated field methodologies to detect and quantify potential CO\(_2\) leakage from geologic storage sites.

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References


Table 1. Properties of two-dimensional transverse model of shallow release.

<table>
<thead>
<tr>
<th></th>
<th>Soil</th>
<th>Cobble</th>
</tr>
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<tr>
<td>Porosity</td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td>Permeability</td>
<td>$5 \times 10^{-11}$ m$^2$</td>
<td>$3.2 \times 10^{-12}$ m$^2$</td>
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<td>Capillary Pressure</td>
<td>$\lambda = 0.291, S_o = 0.15, \alpha = 2.04 \times 10^4$ Pa$^{-1}, P_{\text{max}} = 5 \times 10^5$ Pa, $S_l = 1$.</td>
<td>$\lambda = 0.627, S_o = 0.10, \alpha = 1.48 \times 10^3$ Pa$^{-1}, P_{\text{max}} = 5 \times 10^5$ Pa, $S_l = 1$.</td>
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<tr>
<td>Relative permeability</td>
<td>$S_o = 0.17, S_{og} = 0.05$</td>
<td>$S_o = 0.12, S_{og} = 0.05$</td>
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<td>Molec. diffusivity</td>
<td>Liquid: $10^{-10}$ m$^2$ s$^{-1}$, Gas: $10^{-5}$ m$^2$ s$^{-1}$</td>
<td>Liquid: $10^{-10}$ m$^2$ s$^{-1}$, Gas: $10^{-5}$ m$^2$ s$^{-1}$</td>
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$^1$Pruess et al., 1999.
Figure Captions

**Figure 1.** (a) Surface and horizontal well elevation. Black squares are packers numbered 0-6. Plots of $F_{\text{CO}_2}$ measured along the surface well trace on (b) 17-18 July 2007 and (c) 7-8 August 2007. Distance = 0 m corresponds to grid origin shown in Figure 2.

**Figure 2.** Log $F_{\text{CO}_2}$ maps for measurements made on (a-j) 7-16 July 2007 and (k-n) 9-12 August 2007. Black dots show measurement locations. White circles show grid origin. White line in (a) shows approximate surface well trace.

**Figure 3.** Plot of CO$_2$ discharge versus time for Releases 1 and 2. $D_{\text{tot}}$ (black dots), $D_{\text{back}}$ (open circles), and $D_{\text{leak}}$ (black squares) are total, background (soil respiration), and leakage discharges, respectively. Black line shows simulated time evolution of leakage CO$_2$ flux directly over well.

**Supplement 1.** (a) Surface profile of simulated leakage CO$_2$ flux across well and (b) corresponding cross-section of simulated subsurface CO$_2$ concentrations (mass fraction in the gas phase) for Release 1, Day 8. Black circle is cross section of horizontal well and white lines are contours of liquid saturation (contour interval = 0.2). (c) Surface profile of simulated leakage CO$_2$ flux across well and (d) corresponding cross-section of subsurface CO$_2$ concentrations for Release 2, Day 8.
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