

LEAKAGE AND SEEPAGE IN THE NEAR-SURFACE ENVIRONMENT: AN INTEGRATED APPROACH TO MONITORING AND DETECTION

Curtis M. Oldenburg and Jennifer L. Lewicki

Earth Sciences Division 90-1116, LBNL, Berkeley CA 94720

Abstract

Monitoring and detection of leakage and seepage of carbon dioxide (CO₂) in the near-surface environment is needed to ensure the safety and effectiveness of geologic carbon sequestration. Large leakage fluxes, e.g., through leaking wells, will be easier to detect and monitor than slow and diffuse leakage and seepage. The challenge of detecting slow leakage and seepage is discerning a leakage or seepage signal from within the natural background variations in CO₂ concentration and flux that are controlled by a variety of coupled processes in soil. Although there are no direct examples of leaking geologic carbon sequestration sites on which to base a proposed verification approach, we have been guided by our prior simulation studies of CO₂ leakage and seepage, which showed that large CO₂ concentrations can develop in the shallow subsurface even for relatively small CO₂ leakage fluxes. A variety of monitoring technologies exists for measuring CO₂ concentration and flux, but there is a gap between instrument performance and the detection of a leakage or seepage signal from within large natural background variability. We propose an integrated approach to monitoring and verification. The first part of our proposed approach is to characterize and understand the natural ecosystem before CO₂ injection occurs so that future anomalies can be recognized. Measurements of natural CO₂ fluxes using accumulation chamber (AC) and eddy correlation (EC) approaches, soil CO₂ concentration profiles with depth, and carbon isotope compositions of CO₂ are needed to characterize the natural state of the system prior to CO₂ injection. From this information, modeling needs to be carried out to enhance understanding of carbon sources and sinks so that anomalies can be recognized and subject to closer scrutiny as potential leakage or seepage signals. Long-term monitoring using AC, EC, and soil-gas analyses along with ecosystem and flow and transport modeling should continue after CO₂ injection. The integrated use of multiple measurements and modeling offers a promising approach to discerning and quantifying a small CO₂ leakage or seepage signal from within the expected background variability.

Introduction

One of the outstanding challenges of geologic carbon sequestration is verification, that is, ensuring that carbon dioxide (CO₂) is not leaking from the intended sequestration formation and seeping out of the ground. The most straightforward way of verifying CO₂ sequestration would seem to be direct monitoring and detection of anomalous CO₂ in the near-surface environment. While catastrophic releases to the atmosphere, such as through well blowouts, will be obvious failures and therefore present no challenge for detection, slow or diffuse leakage and seepage of CO₂ will be much more difficult to detect, monitor, and quantify. The difficulty of observing and quantifying diffuse CO₂ leakage and seepage arises because there are large spatial and temporal changes in CO₂ concentration and flux in natural ecosystems, making the main challenge the detection of a CO₂ leakage or seepage signal from within the natural background variation. We have developed an approach for monitoring and verification that involves a variety of integrated measurements and modeling that could be used to discern a CO₂ leakage or seepage signal.

In this brief paper, we summarize our proposed approach for geologic carbon sequestration verification. This approach is guided by results of numerical simulations of CO₂ leakage and seepage that we have carried out over the last few years, and by our experience in monitoring natural systems. Next we review controls on natural CO₂ in the shallow subsurface, and the technologies used for detecting and monitoring CO₂ in the near-surface environment. Finally, we present our ideas for an integrated approach to CO₂ verification.

*Corresponding author: Tel. (510) 486-7419, Fax. (510) 486-5686, Email: cmoldenburg@lbl.gov

Review of Simulated CO₂ Leakage and Seepage

In prior work, we carried out simulations of CO₂ leakage and seepage using T2CA, a recently developed extension of TOUGH2 [1] that models CO₂ migration and dispersion in the subsurface and atmospheric surface layer. Details of the methods used in T2CA can be found in Oldenburg and Unger [2,3]. Numerical simulations showed that CO₂ concentrations can build up to high levels in the vadose zone (~100% CO₂ in soil gas) even when the leakage occurs at a relatively small rate. The reason for this is that there are very few dissipative processes for leaking CO₂ in the vadose zone. Examples of dissipative processes are dissolution of CO₂ into vadose zone moisture, and barometric pumping. However, even for relatively slow leakage rates (e.g., $\sim 10^4$ kg yr⁻¹), these processes do not significantly attenuate CO₂ leakage in the vadose zone [2]. Although the shallow subsurface CO₂ concentrations can be high, the CO₂ seepage flux across the ground surface can be small as CO₂ emissions occur over a large area. Our simulation results for the atmospheric surface layer show that CO₂ concentrations are very low above the ground surface, due to wind and turbulence which dilute and disperse CO₂ [3]. As wind speed increases, the surface-layer concentrations diminish to small values above the background concentration of 370 ppmv [3]. These observations of the expected behavior of leaking and seeping CO₂ have led us to focus monitoring and verification on the shallow subsurface, where CO₂ concentrations will tend to be high, and the daily and seasonal variability will be diminished relative to the above-ground region.

Natural Background CO₂ Fluxes and Concentrations

Background CO₂ in soil is derived primarily from the atmosphere and respiration, i.e., biologically mediated oxidation of organic carbon. A diagram of soil CO₂ sources and exchanges is shown in Figure 1. As shown in Figure 1, background soil CO₂ fluxes and concentrations are dependent on exchange with the atmosphere, production from decay of organic matter such as leaf litter, uptake by plants, production by root respiration, deep degassing, release from groundwater due to depressurization, and production by oxidation of organic carbon in groundwater at the water table. These processes are dependent on temperature, moisture, soil aeration, biological activity, and other factors [4]. The CO₂ degassed by flowing deep groundwater can have its CO₂ source from shallower sources (e.g., respiration, atmospheric CO₂, or particulate organic carbon) encountered at shallower depths over the long groundwater migration pathway (e.g., [5]). In summary, many complex and interrelated processes are active in controlling the natural background variation of CO₂ concentration, flux, and isotopic composition, and these processes can vary on short (hourly) and much longer time scales.

Near-Surface Monitoring Technologies

A wide range of methods is available for monitoring and detection of CO₂ fluxes and concentrations [6,7]. The infrared gas analyzer (IRGA) is used to measure CO₂ concentrations and is based on the principle of light absorption in the near-infrared part of the spectrum, typically 4.26 μ m. IRGA-based devices are inexpensive, portable, and can measure CO₂ over a wide range of concentrations. Promising new methods of CO₂ monitoring include light detection and range finding (LIDAR), and new approaches such as hyperspectral imaging of vegetative stress [8].

Two basic approaches exist for measuring CO₂ fluxes: (1) the accumulation chamber (AC), and (2) the eddy correlation (EC) approach. In the AC method, an open-bottomed chamber is placed directly on the soil surface or on a collar installed on the ground surface and the rate of soil-CO₂ accumulation is measured using an IRGA (Figure 2a). This approach provides a small-scale measurement of soil-CO₂ flux. The measurement is relatively quick and many such measurements can be made over a large area, with statistical approaches used to interpolate an overall areal flux [7]. The EC method provides a spatially averaged flux by correlating CO₂ concentration measured at a fixed height above the ground using an IRGA with local meteorological variations at the same elevation (Figure 2b). After time-averaging of the local variations of concentration and vertical wind speed, an average flux over a given footprint is derived. The footprint area is a function of the instrument height above the ground surface and local wind velocity, and is on the order of 10–100 times the instrument height. The advantage of EC is that it provides a spatially averaged flux that includes plant and soil sources; the limitation is that it assumes a horizontal ground surface and uniform plant cover over the footprint [9].

Proposed Integrated Approach to Sequestration Verification

There is a wide gap between the performance capabilities of technological devices and their applicability for geologic carbon sequestration leakage or seepage monitoring, detection, and verification. The fundamental challenge for verification when leakage and seepage fluxes are small is discerning the CO₂ leakage or seepage signal from natural background variability. Because CO₂ leakage and seepage at the ground surface is expected to be a very rare phenomenon, monitoring at CO₂ sequestration sites will typically record background natural variability of the ecological system. To meet the challenge of CO₂ sequestration verification, sophisticated procedures or new technologies may have to be developed. In this section, we discuss ways that the CO₂ leakage or seepage signal could be discerned and quantified from measurements made in the near-surface environment with existing conventional devices.

The approach we suggest is based on our experience in modeling and field measurements of CO₂ emissions [2,3,10]. Experience has shown that the subsurface, even at depths of less than a meter, is much less affected by surface environmental processes and the strong dispersion effects of surface-layer winds and barometric pressure changes. Therefore, we expect the changes in CO₂ concentration, flux, and isotopic composition to be most recognizable in the subsurface, and therefore we focus our attention on monitoring and detection in the shallow subsurface. A great deal of effort should be put into understanding the natural ecological system and its baseline properties prior to CO₂ injection so that future anomalies in CO₂ concentration and flux can be identified. Particular attention should be given to understanding the natural variability in areas where leakage or seepage may be expected, e.g., near faults or around wells. Modeling and analyses of expected ecological (e.g., [11]) and CO₂ transport dynamics (e.g., [1,2,3]) should be integrated with the measurements to develop a comprehensive understanding of the system with and without CO₂ leakage and seepage.

We propose a variety of measurements for CO₂ sequestration verification. First, bulk soil-gas composition can be analyzed as a function of depth (i.e., vertical soil-gas profiles). Increasing CO₂ concentrations with depth indicate a CO₂ source at depth. If the source is oxidative decay of organic matter, there should be an associated decrease in O₂ concentration, the absence of which may suggest a CO₂ leakage source. A more definitive way of distinguishing the source of carbon in CO₂ in soil is by isotopic analyses. The characteristic properties of gas composition profiles and isotopic signatures for different sources of CO₂ are summarized in Table 1. Both the stable ¹³C and radiogenic ¹⁴C vary depending on the source of CO₂. However, only the absence of ¹⁴C coupled with high local CO₂ concentrations provide a strong indicator of a fossil-fuel origin of CO₂ consistent with leaking CO₂ from a sequestration site. The overlap in ¹³C values from the various potential sources of CO₂ makes ¹³C less useful as a leakage indicator. A large number of measurement locations will be required and statistical approaches will have to be used to interpolate relatively sparse measurements over the area of interest. In addition to measurements of gas composition and variation with depth, seepage flux should be measured using either AC or EC approaches. Areas of anomalous emissions would be obvious places to focus additional sampling and monitoring efforts. Measurements of gas components other than CO₂ associated with the CO₂ sequestration target, e.g., CH₄ if the sequestration target formation is a hydrocarbon reservoir, may also be a promising indicator of potential leakage [12].

Integrated with both the baseline (i.e., pre-injection) characterization measurements and the ongoing monitoring should be numerical simulation [1,2,3] and ecological modeling [e.g., 11]. These analysis efforts should both make use of the measured observations (e.g., as calibration data) as well as make predictions that can be compared against the measurements, with disparities becoming the focus of particular scrutiny as potential leakage or seepage signals. In Figure 4, we present a proposed schedule of integrated monitoring and modeling activities for verification. We emphasize the need for pre-injection activities along with long-term monitoring during and after injection.

Conclusions

Verification of geologic carbon sequestration by monitoring and modeling in the near-surface environment is a challenging but necessary task to ensure the safety and effectiveness of geologic carbon sequestration. The challenge arises because of the large natural background variability in CO₂ concentration and flux. We propose an approach that integrates several different measurements with modeling and analysis. Discrepancies between

measurements and model results should be analyzed closely as they may point to evidence of CO₂ leakage or seepage.

Acknowledgments

We thank xxx and xxx for reviews of an earlier draft. This work was supported in part by the Office of Science, U.S. Department of Energy, and by the Assistant Secretary for Fossil Energy, Office of Coal and Power Systems, through the National Energy Technologies Laboratory (NETL), and by Lawrence Berkeley National Laboratory under Department of Energy Contract No. DE-AC03-76SF00098.

References

1. Pruess, K., C. Oldenburg, and G. Moridis. 1999. TOUGH2 User's Guide Version 2.0, Lawrence Berkeley National Laboratory Report *LBL-43134*, 197 pp.
2. Oldenburg, C.M., and A.J.A. Unger. 2003. On leakage and seepage from geologic carbon sequestration sites: unsaturated zone attenuation, *Vadose Zone Journal*, 2, 287–296.
3. Oldenburg, C.M., and A.J.A. Unger. 2004. Coupled vadose zone and atmospheric surface-layer transport of CO₂ from geologic carbon sequestration sites, *Vadose Zone Journal*, in press.
4. Amundson, R.G., O.A. Chadwick, and J.M. Sowers. 1989. A comparison of soil climate and biological activity along an elevation gradient in the eastern Mojave Desert, *Oecologia*, 80, 395–400.
5. Wood, W.W. and M.J. Petraitis. 1984. Origin and distribution of carbon dioxide in the unsaturated zone of the southern high plains of Texas, *Water Resour. Res.*, 20, 1193–1208.
6. Shuler, P.J. and Y. Tang. 2002. *Atmospheric CO₂ Monitoring Systems – A Critical Review of Available Techniques and Technology Gaps: Report for SMV Group*, The CO₂ Capture Project (CCP).
7. Oldenburg, C.M., and J.L. Lewicki. 2003. Near-Surface Monitoring Strategies for Geologic Carbon Dioxide Storage Verification, Lawrence Berkeley National Laboratory Report *LBL-54089*.
8. Martini, B.A., D.C. Potts, E.A. Silver and W.L. Pickles. 2000. Hyperspectral remote sensing for research and monitoring in active volcanic regions: Long Valley Caldera, CA, *EOS Trans. Am. Geophys. Union*, Fall Meeting, p. F1385.
9. Baldocchi, D.D., R. Valentini, S. Running, W. Oechel, and R. Dahlman. 1996. Strategies for measuring and modeling carbon dioxide and water vapor fluxes over terrestrial ecosystems, *Global Change Biology*, 2, 159–168.
10. Lewicki, J.L., W.C. Evans, G.E. Hilley, M.L. Sorey, J.D. Rogie, and S.L. Brantley. 2003. Shallow soil CO₂ flow along the San Andreas and Calaveras faults, California, *J. Geophys. Res.*, 108, ECV 3-1 – ECV 3-14.
11. Bonan, G.B.. 1996. A land surface model (LSM version 1.0) for ecological, hydrological, and atmospheric studies: Technical description and user's guide, *TN-417+STR*, NCAR, Boulder, CO.
12. Klusman, R.W.. 2003. Evaluation of leakage potential from a carbon dioxide EOR/sequestration project, *Energy Conversion and Management*, 44, 1921–1940.

Table 1. Chemical and isotopic signatures related to CO₂ derived from different sources.

CO ₂ source	$\delta^{13}\text{C}_{\text{CO}_2}$ ‰	$\Delta^{14}\text{C}_{\text{CO}_2}$ ‰	Near-surface CO ₂ conc.	CO ₂ conc. depth profile	O ₂ conc. depth profile
Atmosphere	-7	-70	Low	–	–
Plant root respiration and oxidative decay of young soil organic matter	C ₃ : -24 to -38 C ₄ : -6 to -19	≥ -70	Low to moderate	Increasing through soil zone	Decreasing through soil zone
Oxidative decay of ancient organic matter	C ₃ : -24 to -38 Aquatic/C ₄ : -6 to -19 Also age dependent	Highly depleted to absent, depending on age	Low	Increasing potentially through vadose zone	Decreasing potentially through vadose zone
Marine carbonate rocks	0 ± 4	absent	Low	Increasing through vadose zone	No effect
Fossil fuel	Average: -27	absent	Moderate to high	Increasing through vadose zone	No effect

Conc., C₃, and C₄, refer to concentration, C₃ plants, and C₄ plants. All near-surface concentrations given are general estimates; these concentrations will be strongly dependent on the magnitude of the CO₂ flux.

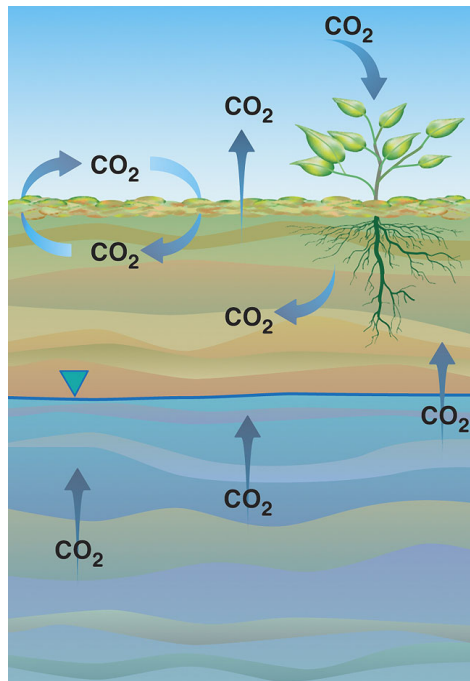


Figure 1. Soil CO₂ sources and sinks, showing from left-right, top-bottom, exchange with the atmosphere, production from decay of organic matter such as leaf litter, uptake by plants, production by root respiration, deep degassing, release from groundwater due to depressurization, and production by oxidation of organic carbon in groundwater at the water table.

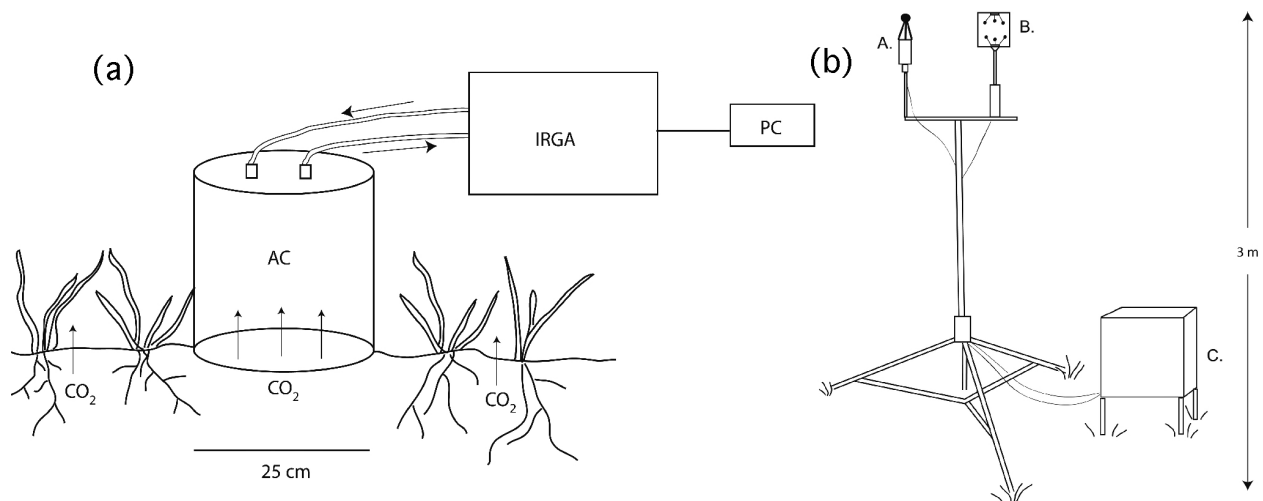


Figure 2. Sketch of (a) accumulation chamber (AC) measurement system of soil CO₂ flux in which contained air is circulated through the AC and the IRGA and the rate of change of CO₂ concentration in the AC is measured by the IRGA and recorded by the PC, and (b) eddy correlation (EC) instrumentation tower showing (A) open-path IRGA, (B) high frequency response sonic anemometer, and (C) box containing power source and datalogger/PC.

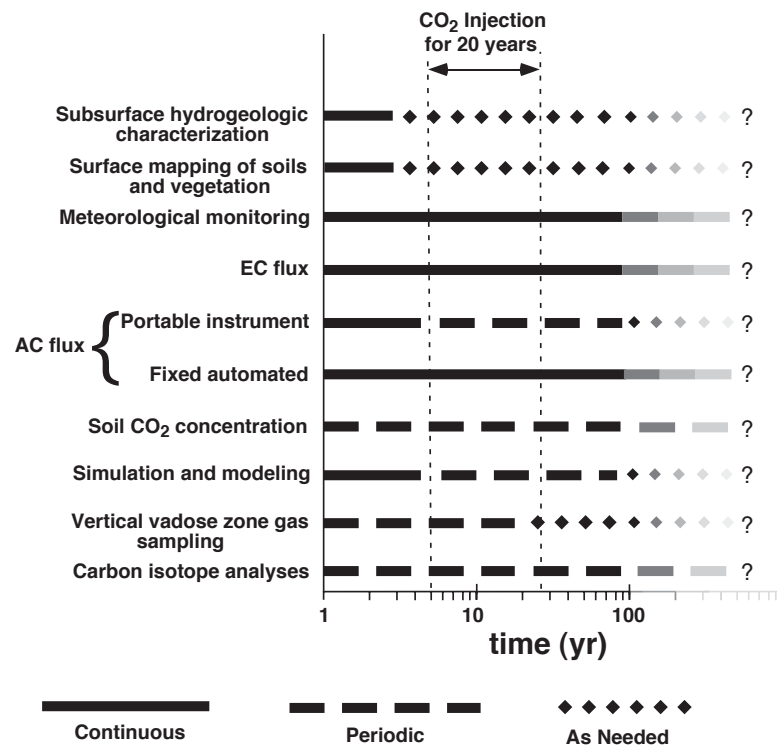


Figure 3. Example activity and schedule chart for CO₂ leakage or seepage signal detection and monitoring showing generalized frequency of measurements (i.e., continuous, periodic, as needed) over time preceding, during, and following CO₂ injection. Lighter shading indicates increasing uncertainty in need for activities at long times following injection.