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Rapid detection and characterization of surface CO2 leakage through the real-time measurement of 13C signatures in CO2 flux from the ground

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The surface monitoring of CO_2 over geologic sequestration sites will be an essential tool in the monitoring and verification of sequestration projects. Surface monitoring is the only tool that currently provides the opportunity to detect and quantify leakages on the order of 1000 tons/year CO_2. Near-surface detection and quantification can be made complicated, however, due to large temporal and spatial variations in natural background CO_2 fluxes from biological processes. In addition, current surface monitoring technologies, such as the use of IR spectroscopy in eddy covariance towers and aerial surveys, radioactive or noble gas isotopic tracers, and flux chamber gas measurements can generally accomplish one or two of the necessary tasks of leak detection, identification, and quantification, at both large spatial scales and high spatial resolution. It would be useful, however, to combine the utility of these technologies so that a much simplified surface monitoring program can be deployed.

Carbon isotopes of CO_2 provide an opportunity to distinguish between natural biogenic CO_2 fluxes from the ground and CO_2 leaking from a sequestration reservoir that has ultimate origins in a process giving it a distinct isotopic signature such as natural gas processing. Until recently, measuring isotopic compositions of gases was a time-consuming and expensive process utilizing mass-spectrometry, not practical for deployment in a high-resolution survey of a potential leakage site at the surface. Recent developments in commercially available instruments utilizing wavelength scanned cavity ringdown spectroscopy (WS-CRDS) and Fourier transform infrared spectroscopy (FT-IR) have made it possible to rapidly measure the isotopic composition of gases including the ^{13}C and ^{12}C isotopic composition of CO_2 in a field setting.

A portable stable carbon isotope ratio analyzer for carbon dioxide, based on wavelength scanned cavity ringdown spectroscopy, has been used to rapidly detect and characterize an intentional leakage of CO_2 from an underground pipeline at the ZERT experimental facility in Bozeman, Montana. Rapid (1 hour) walking surveys of the entire 100m x 100m site were collected using this mobile, real-time instrument. The resulting concentration and ^{13}C isotopic abundance maps were processed using simple yet powerful analysis techniques, permitting not only the identification of specific leakage locations, but providing the ability to distinguish petrogenic sources of CO_2 from biogenic sources.

At the site an approximately 100-meter horizontal well has been drilled below an alfalfa field at a depth between 1-3 meters below the surface. The well has perforations along the central 70 meters of the well. The overlying strata are highly permeable sand, silt, and topsoil. For 30 days starting July 15, 2009, CO_2 was injected at a rate of 0.2 tonnes per day. The injection rate is designed to simulate leakage from a mature storage reservoir at an annual rate of between .001 and .01%. The isotopic composition of the gas from the tank is at $\delta^{13}C$ signature of approximately -52 parts per thousand (per mil), far more negative than either atmospheric (approx. -8 per mil) or CO_2 from soil respiration (approx. -26 per mil) at the site. The CO_2 isotopic and concentration measurements were taken with a Picarro WS-CRDS analyzer with 1/8" tubing connected to a sampling inlet. Simultaneous with CO_2 concentrations (including ^{13}C), position data was logged using a GPS receiver. Datapoints are taken around every second. The analyzer was powered using batteries and housed in a mobile cart.

The surveys consisted of traverses of the site along the length of the pipeline and extending out 100 meters on either side of the pipeline with the end of the gas inlet tube approximate 9 cm above the ground at a speed of 1-2m/sec. This simulates the type of survey that could be easily performed if the actual or potential site of a leak was known to within an area on the order of 100 square kilometers or less, the scale of expected industrial CO_2

sequestration operations. The surveys were performed both during the day and during the evening when CO_2 flux due to respiration from the soil is markedly different.

Keeling plots were used to characterize the spatially varying ^{13}C composition of ground source CO_2 across the site. A map constructed from this data shows that CO_2 flux from sources of leakage was characterized by a $\delta^{13}C$ of -40 per mil or less whereas locations away from the leakage spots had much higher $\delta^{13}C$ signatures, -25 per mil or higher.

The distinct isotopic signature allows for a clear discernment between leakage of petrogenic CO_2 and that of natural CO_2 fluxes from soil respiration. This is particularly valuable in the circumstance where the leak is slow enough that it could not be identified from CO_2 concentration or flux changes above the natural background signal alone. Moreover, this detection took place both rapidly and at high spatial resolution. Samples collected from a mobile platform moving at the rate and with the sampling frequency used in this study could provide a 1000 km of survey traverses over an area of 100 km^2 within 2-3 weeks. This provides a powerful tool for surface monitoring, combining the utilities of leak detection, characterization, and source identification with rapid deployment across large spatial scales and high spatial resolutions.