ZERT Controlled Release Experiment

Lee H. Spangler Director, ZERT and The Energy Research Institute Montana State University

Zero Emissions Research & Technology

A collaborative involving Universities and DOE National Labs

- Montana State University Lead Institution
- Los Alamos National Laboratory
- Pacific Northwest National Laboratory
- West Virginia University
- Lawrence Berkeley National Laboratory
- National Energy Technology Laboratory
- Lawrence Livermore National Laboratory





Montana - A Brief Comparison





<u>Japan</u>

Area: Population

377,930 km² 127,078,680

Germany

Area: 357,021 km² Population 82,282,988 0.7% of world's coal reserves

Norway

Montana

Population

6% of world's coal reserves

Significant Oil & Gas

Area:

Area: Population 3rd largest oil exporter

384,802 km² 4,660,539

967, 440







Atmosphere

Biosphere Soil (Vadose & Shallow Saturated Zones)

Caprock & Deep Overburden

Injection Zone





Near – Surface Monitoring Zones

- Atmosphere
 - Ultimate Integrator
 - Dynamic
 - Monitoring & Modeling
- Biosphere
 - dynamic
 - requires protection
 - opportunity for wide area monitoring but indirect methods
- Soil
 - Integrates
 - dynamic
- Aquifers
 - Integrates
 - Requires protection



Motivation (2006)

The situation in 2006 when we started planning the work:

- Near-surface detectors were considered highly desirable for public assurance
- They had been deployed at sequestration pilot sites
- These pilot sites were well chosen and do not leak
- Thus, the near-surface detection techniques had not been adequately tested under realistic conditions
- The primary initial purpose was detection verification





Facility Goals

- Develop a site with known injection rates for testing near surface monitoring techniques
- Use this site to establish detection limits for monitoring technologies
- Use this site to improve models for groundwater vadose zone – atmospheric dispersion models
- Develop a site that is accessible and available for multiple seasons / years





Scaling



Imagine a realistic feature that might result in leakage

We chose to mimic a fault which might be on the order of 1 km long with a surface expression of 10m – 100m in width.

A 100m horizontal well would be 10% of the first case and 1% of the second case.

Scaling by a factor of 10 – 100 is a reasonable extrapolation

Sally Benson Lee Spangler





What Are Relevant Release Rates?

- 4 Mt/year injection from 500 MW power plant
- 50 years injection Total of 200 Mt Injected
- Consider maximum leakage rates discussed to mitigate climate change

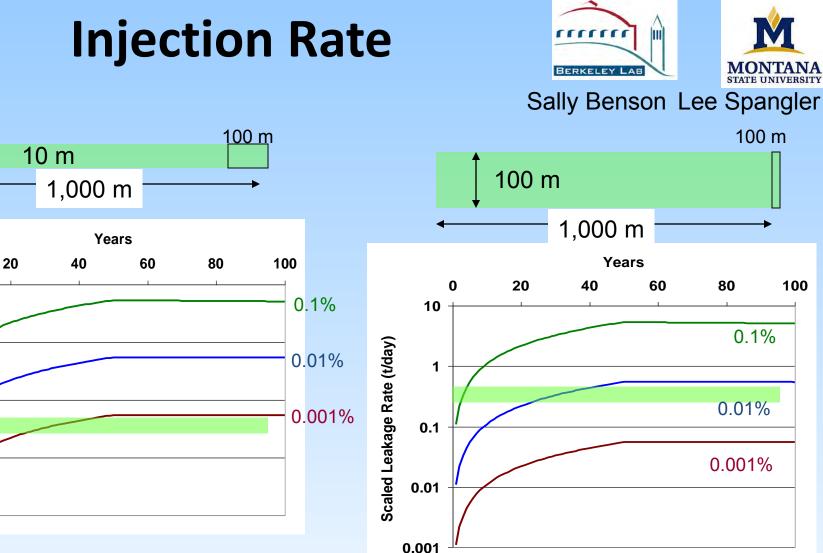
- 1% over 100 years = 0.01% / year = 0.0001

- 1% over 1000 years = 0.001% / year = 0.00001

- 200,000,000 x 0.00001 = 2,000 Tonnes / yr
- 5.5 Tonnes / Day
- This is the equivalent of about 85 idling cars







Scale to 1000 m leak 1,000 kg/day: 1 tonne/day

1

0

100

10

1

0.1

0.01

Scaled Leakage Rate (t/day)

We used a 0.15 Tonne / Day rate

An idling car generates about 0.04545 kg CO_2 / min or 64.5 kg CO_2 / day. Our injection rate is about equal to 2.3 idling cars





Field Test Facility







EST 1943

🤟 WestVırginiaUniversity.

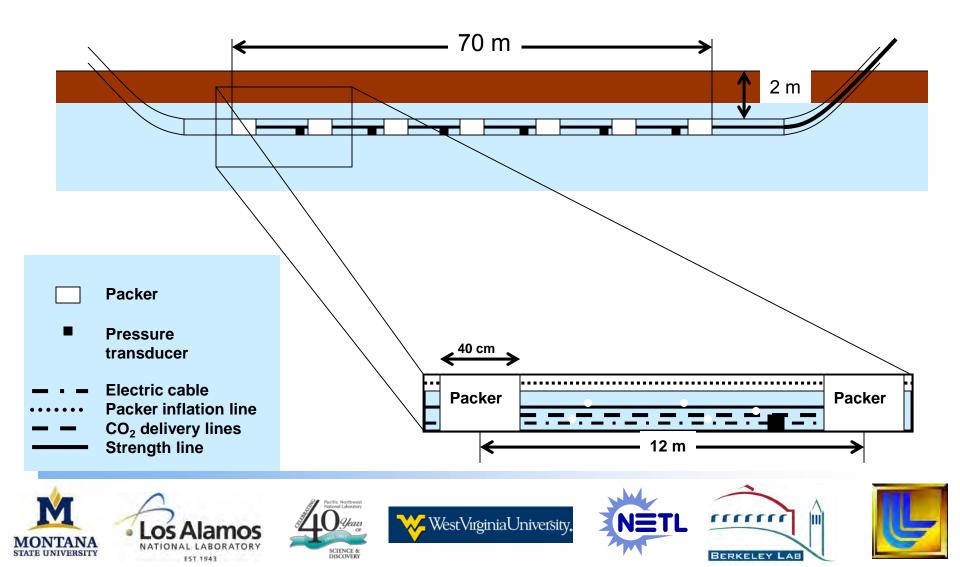


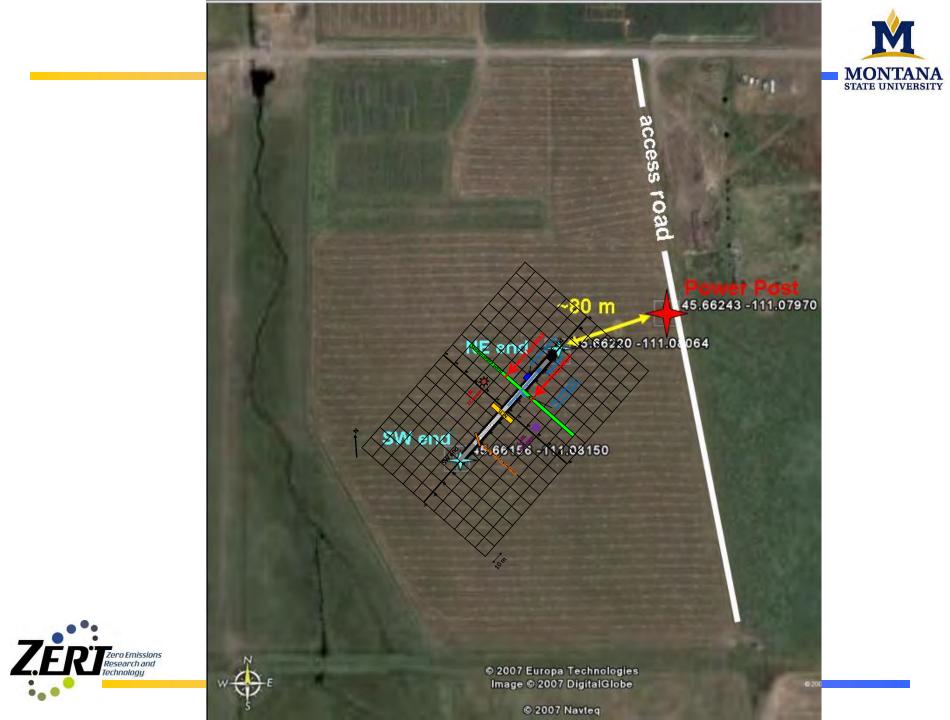






Ray Solbau, Sally Benson





Methods

- Soil Gas Monitoring
- In-situ soil gas probes
- Eddy Covarience
- Soil Flux chambers
- Differential Absorption LIDAR
- Cavity ring-down, other isotopic measurements
- Water chemistry
- Tracers
- Hyperspectral / mutispectral imaging
- Many more

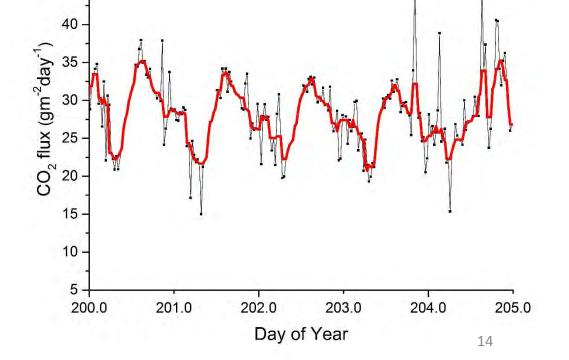




CO₂ Background is Highly Variable



Affected by sunlight, precipitation, wind, etc. Red line shows diurnal variation, but there are also short term and much longer term variations



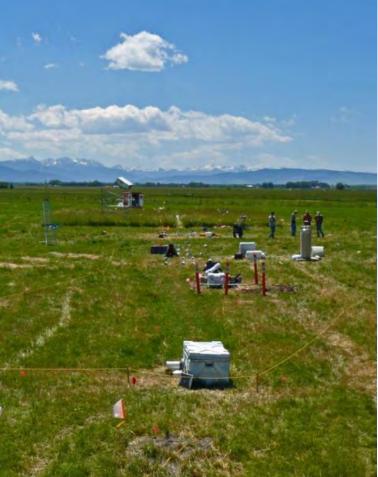
raw flux data

smoothed flux data



Large Number of Participants / Methods

47 investigators31 instruments / sensor arrays5 univ. 6 DOE labs, 4 companies



Investigator	Institution	Monitoring Technology	Number of Sensors
Arthur Wells Rod Diehl Brian Strasizar	National Energy Technology Laboratory	Atmospheric tracer plume measurements	1 tower (4m) Blimp (Apogee Scientific) with 3 tether line samplers
		Bee hive monitoring for tracer with sorption tube and pollen trap	2 hives
		Automated Soil CO ₂ flux system	4 chambers
William Pickles Eli Silver Erin Male	University of California- Santa Cruz	Hand held hyperspectral measurements (plant health)	1 instrument
Yousif Kharaka James ThordsenGil AmbatsSarah Beers	United States Geological Survey*	Ground water monitoring	1 EC and temperature probe, Dissolved oxygen probe, lab analysis of water samples
Henry Rauch	West Virginia University	Water monitoring well headspace gas sampling	1 sensor
Lucian Wielopolski Sudeep Mitra	Brookhaven National Laboratory*	Ineleastic neutron scattering (total soil carbon)	1 instrument
Martha Apple Xiaobing Zhou Venkata Lakkaraju Bablu Sharma +2 students	Montana Tech*	Soil moisture, temp. Chlorophyll Content Meter , Fluorescence Meter , LI-COR 2000 to measure leaf area index Leaf Porometer to measure stomatal conductance	5 sensors
		Infrared radiometry (plant health) Atmospheric humidity	2 instruments 1 sensor each
		and temperature, accumulated rainfall	
		Plant root imaging	1 camera
		Soil conductivity	1 sensor
		Handheld hyperspectral measurements (plant health)	1 instrument
William Holben Sergio Morales	University of Montana*	Microbial studies	Lab analysis



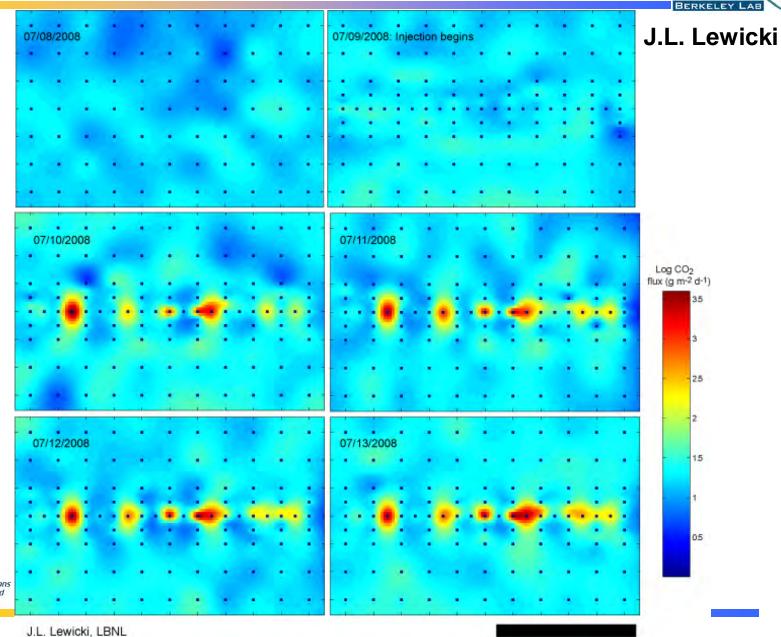
Large Number of Participants / Methods



ZĖRI	Zero Emissions Research and Technology

Investigator	Institution	Monitoring	Number of Sensors
L G 1		Technology	1.5
Lee Spangler	Montana State	Water content	15 sensors
Laura Dobeck	University	reflectometers (soil	
Kadie Gullickson		moisture)	- /
		Automated soil CO ₂	5 long term
		flux system	chambers, 1 portable
			survey chamber
		CO_2 soil gas	6 sensors
		concentration	
Kevin Repasky (PI)	Montana State	Underground fiber	4 sensors
Jamie Barr	University	sensor array (CO ₂ soil	
		gas concentration)	
Rand Swanson	Resonon*	Flight based	linstrument
		hyperspectral	
		imaging system	
Joseph Shaw (PI)	Montana State	Multi-spectral	linstrument
Justin Hogan	University	imaging system (plant	
Nathan Kaufman		health)	
		Meteorological	1 tower
		measurements	
Julianna Fessenden	Los Alamos National	In situ (closed path)	1 instrument
+3 students	Laboratory	stable carbon isotope	
		detection system	
		Flask sampling for in	Lab analysis
		situ isotope detection	
Sam Clegg	Los Alamos National	Frequency-modulated	1 instrument
Seth Humphries	Laboratory	spectroscopy (FMS)	
	×	open-air path	
Thom Rahn	Los Alamos National	Eddy covariance	1 tower
	Laboratory	a 11 ao . a	
James Amonette	Pacific Northwest	Soil CO_2 flux	27 chambers
Jon Barr	National Laboratory	(steady-state)	
Sally Benson (PI)	Stanford University*	Commercial cavity	1 instrument
Sam Krevor	/ Picarro	ringdown real-time	
Jean-Christophe	Instruments*	measurements of $\delta^{13}C$	
Perin		and CO_2 in air	
Ariel Esposito			
Chris Rella (Picarro)	x x .		
Greg Rau	Lawrence Livermore	Commercial cavity	1 instrument
Ian McAlexander	National Laboratory	ringdown real-time	
(LGR)	Los Gatos Research*	measurements of $\delta^{13}C$	
I C I II	I DII	and CO_2 in air	0
Jennifer Lewicki	Lawrence Berkeley	CO ₂ soil gas	8 sensors
	National Laboratory	concentration	
		CO ₂ atmospheric	2 sensors
		concentration	
		Chamber soil CO ₂	1 instrument
		flux measurements	
		Meteorological	1 tower

Flux Chamber





50 m

0

TUDIN

lui)

Eddy covariance net CO₂ flux monitoring

BERKELEY LAB

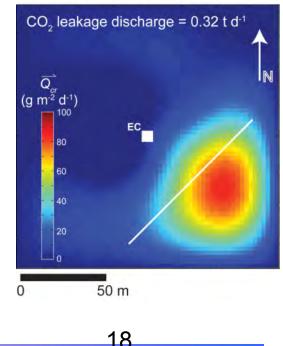
An eddy covariance (EC) station was deployed ~30 m NW of the release well in 2006, 2007, and 2008.

In 2008 (0.3 t $CO_2 d^{-1}$ for 1 month) leakage signal was detected in raw EC CO_2 flux (F_c) data. Ecosystem CO_2 fluxes were modeled and removed from F_c to improve signal detection in residual flux (F_{cr}) data.

Log soil CO2 flux (g m-2 d-1) (a) 90% 0.5 1 1.5 2 2.5 3 3.5 5% g m² d 50% $= -19 \, \text{a} \, \text{m}^2 \, \text{d}$ $\sigma = 32 \text{ g m}^2 \text{ d}^3$ -100 100 (b)F_{ar} (g m-² d⁻¹) 50 m $u = 2 \sigma m^2 d^3$ $\sigma = 15 \, \text{g m}^2 \, \text{d}^2$

J. Lewicki (LBNL)

A least-squares inversion of measured residual CO_2 fluxes and corresponding modeled footprint functions during the 2008 release modeled the distribution of surface CO_2 fluxes, allowing us to locate and quantify (to within 7%) the leakage signal.

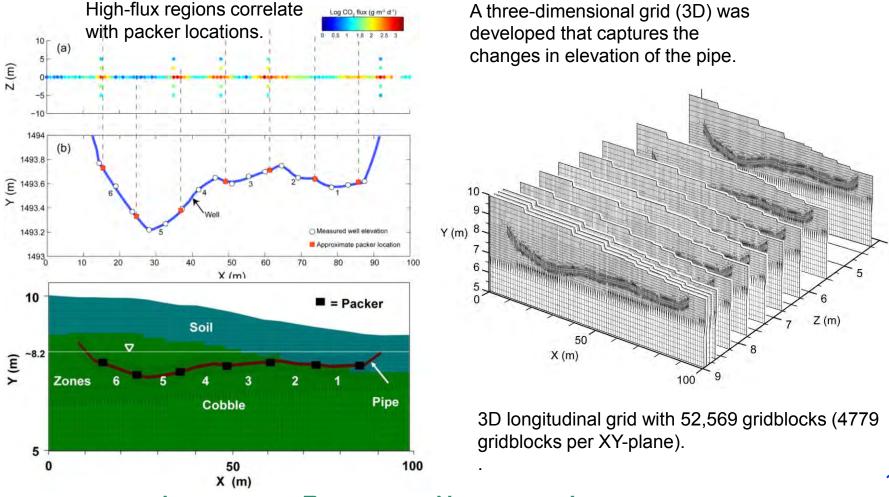




Shallow CO₂ Flow Modeling (1)

C. Oldenburg (LBNL)

TOUGH2/EOS7CA was used to address the origin of patchy emissions at the ZERT shallow-release experiment.



LAWRENCE BERKELEY NATIONAL LABORATORY



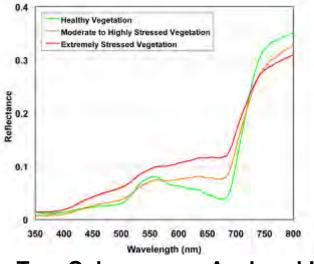
Shallow CO₂ Flow Modeling (2)

C. Oldenburg (LBNL)

Results suggest that packer locations influence emission patterns.

 $q_{CO2} = 100 \text{ kg CO}_2/\text{day}$ Base Case (6 zones) Case 1 (23 zones) Three-dimensional results of X_a^{CO2} Xgcoz at *t* = 3 days showing patchy plane at Z = 8,975 m plane at Z = 8.975 m 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1 emission pattern. (m) Y t = 1.5 hr t = 1.5 hr 20 40 60 80 100 60 80 100 X (m) X (m) Xgcoz plane at Z = 8.975 m plane at Z = 8.975 m 0.9 0.8 0.7 0.6 0.5 0.4 0.3 0.2 0.1 (m) X ۲ (m) t = 12 hr t = 12 hr10 50 100 80 20 40 60 100 X (m) X (m) · Patches are correlated with packer locations and high-۸ (m) 0 elevation regions in each zone in the soil material. 20 + 40 3 60 • With more packers (i.e., more zones), there are still early breakthroughs but overall emission is less patchy. Therefore, simulations support the hypothesis that along-80 pipe flow of CO₂ upwards within each zone leads to an 100 0 10 effective point-source release that creates a persistent Z (m)

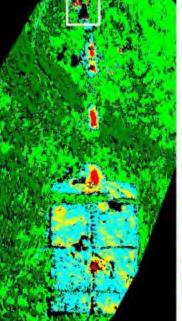
patchy emission.



True Color



Analyzed Image

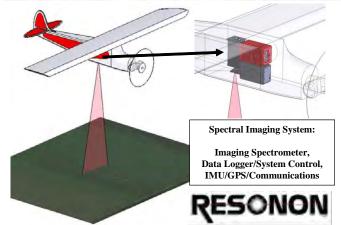


Hyperspectral Imaging









High Stress

Moderate Stress

Low or Seasonal Stress

Healthy Vegetation (Grasses)

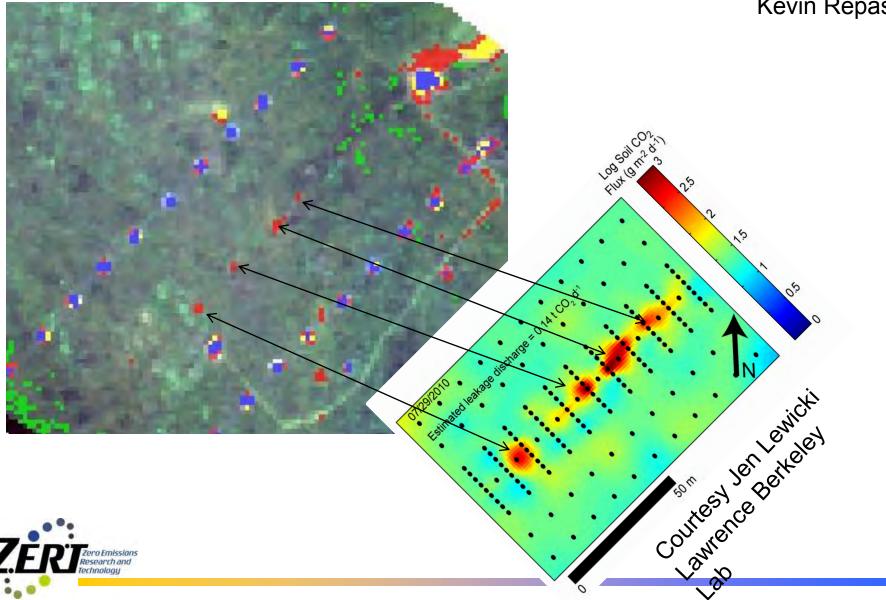
Healthy Vegetation (Herbaceous Legumes)

Unclassified

Hyperspectral Imaging Unsupervised Classification



Kevin Repasky

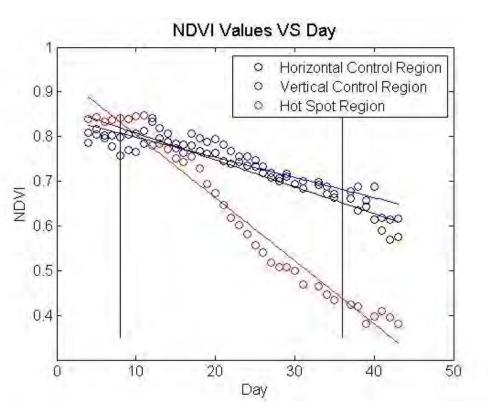




Multi-spectral imaging for detecting CO₂ leaks



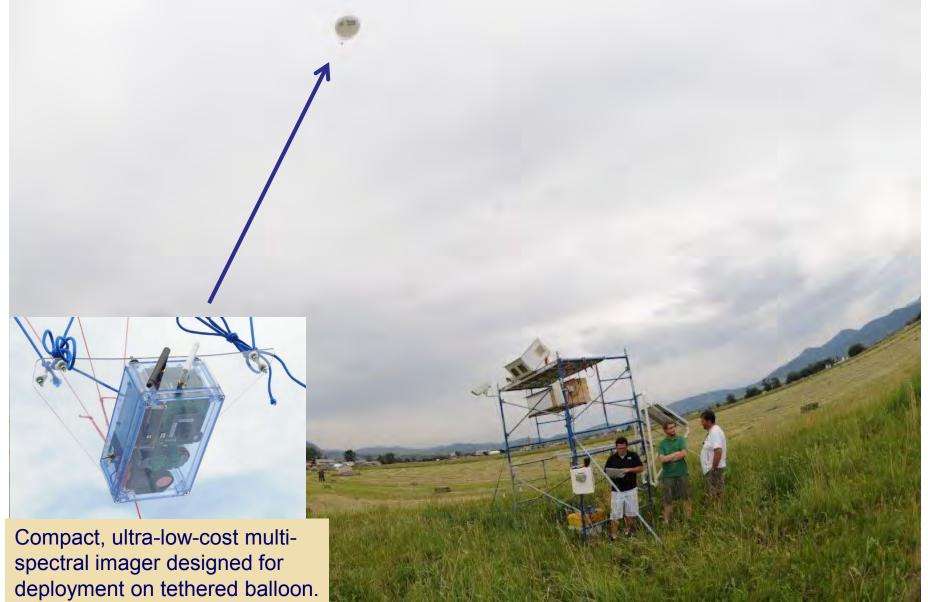
Multispectral imagers used to detect plant stress caused by CO_2 leaking from underground.



Time-series plot showing that the CO₂-affected plant health decays faster over time than the control region. This plot shows Normalized Difference Vegetation Index (NDVI), found from NIR and red reflectances as (NIR-red)/(NIR+red)

Tethered balloon multispectral imaging at ZERT

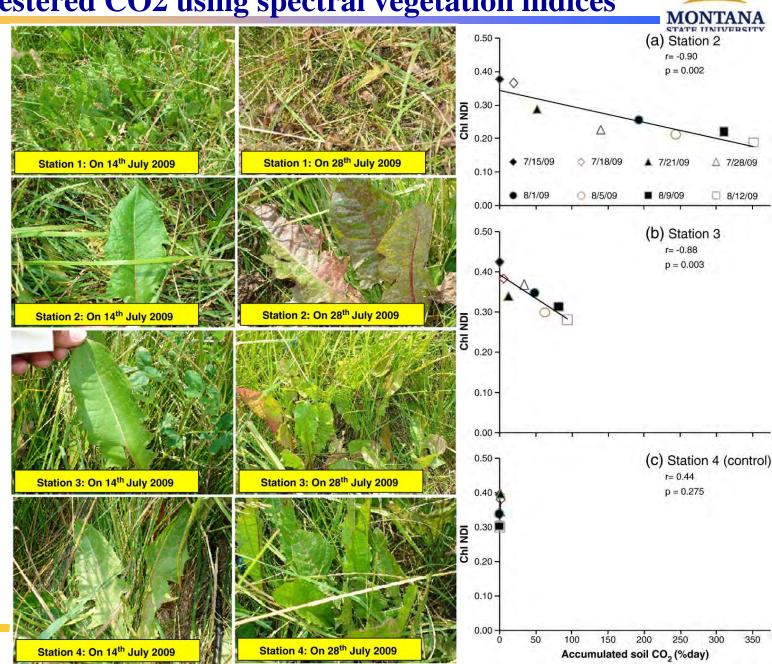
J. Shaw



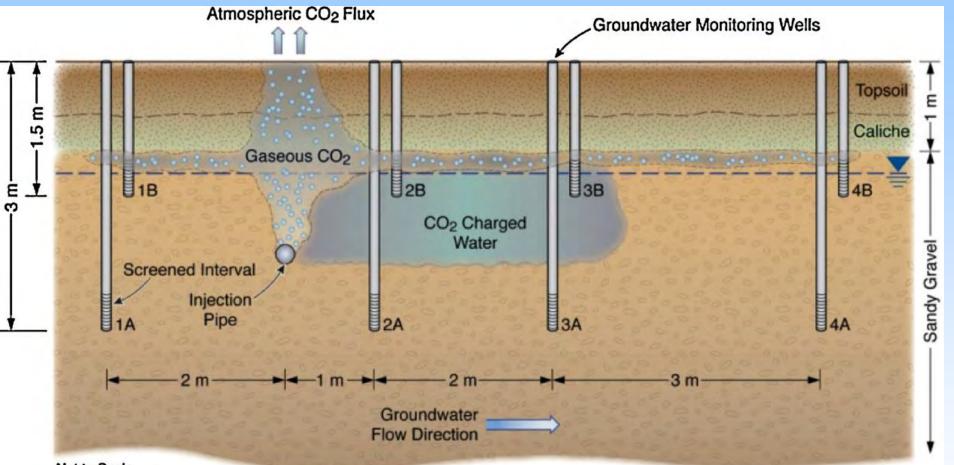
Studying the vegetation response to simulated leakage of sequestered CO2 using spectral vegetation indices 0.50 (a) Station 2 Ecological r= -0.90 p = 0.0020.40 Informatics 5 (2010) 379-389 0.30

Montana Tech

Venkata Ramana Lakkaraju, Xiaobing Zhou, Martha E. Apple, Al Cunningham, Laura M. Dobeck, Kadie Gullickson. Lee H. Spangler



Geochemical Monitoring



Not to Scale



USGS, LBNL, EPRI, WVU, MSU - Environ Earth Sci (2010) 60:273–284 Liange Zheng, John A. Apps, Nicolas Spycher, Jens T. Birkholzer, Yousif K. Kharaka, James Thordsen, Sarah R. Beers, William N. Herkelrath, Evangelos Kakouros, Robert C. Trautz, Henry W. Rauch Kadie S. Gullickson

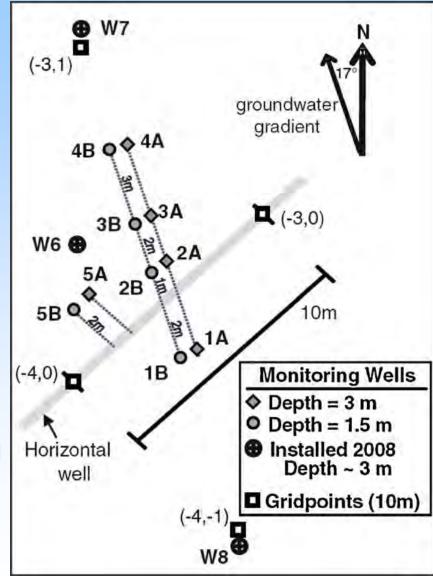
Geochemical Monitoring

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- (1) calcite dissolution could be the primary process buffering pH and releasing Ca+2 in groundwater,
- (2) the increase in the concentrations of major cations and trace metals except Fe could be explained by Ca+2-driven exchange reactions,
- (3) the release of anions from adsorption sites due to competing adsorption of bicarbonate could explain the concentration trends of most anions, and
- (4) the dissolution of reactive Fe minerals (such as fougerite) could explain the increase in total Fe concentration.

Environ Earth Sci (2010) 60:273–284 Int. J. Greenhouse Gas Control (2011)



Atmospheric monitoring of a perfluorocarbon tracer at the 2009 ZERT Center experiment

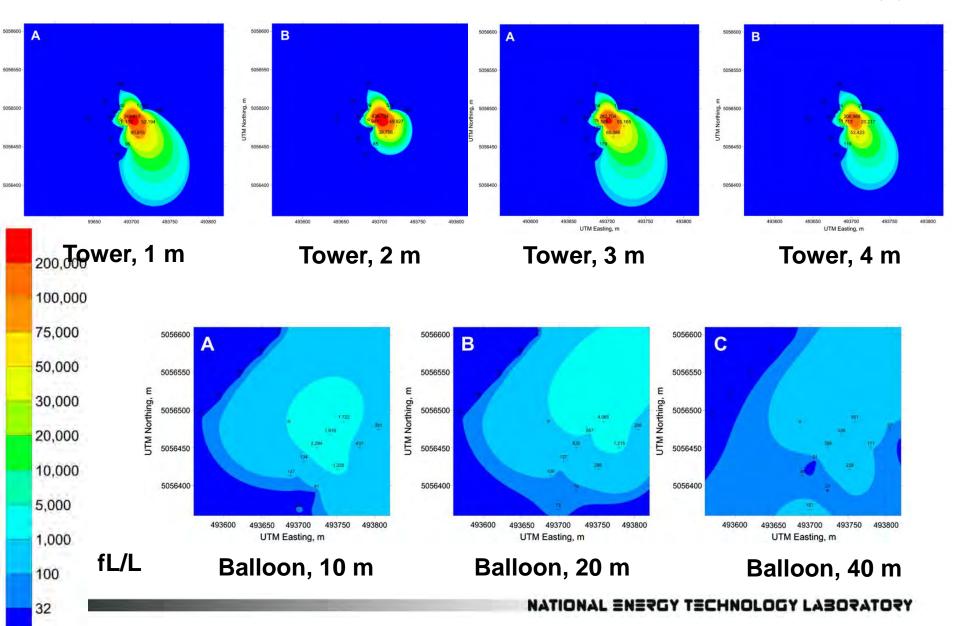


NETL

Natalie Pekney , Arthur Wells , J. Rodney Diehl, Matthew McNeil, Natalie Lesko, James Armstrong, Robert Ference Atmospheric Environment 47 (2012) 124e132

Atmospheric monitoring of a perfluorocarbon tracer

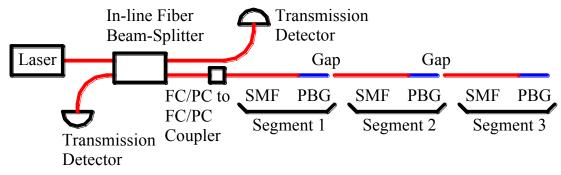






Inline Fiber Sensor

K. Repasky



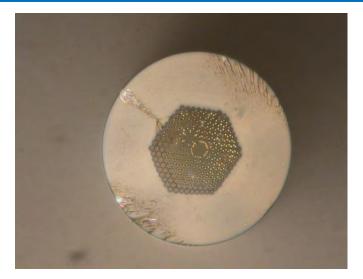
The inline fiber sensor uses a series of segmented photonic bandgap (PBG) fiber in series to for a inline fiber sensor array.

Each segment is addressed using time of flight of the laser pulse.

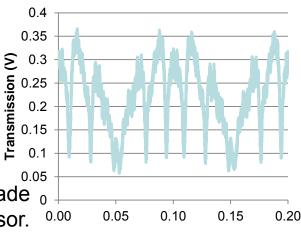
 CO_2 diffuses into the PBG fiber to allow spectroscopic measurements of CO_2 concentration.

Challenge: PBG fiber is larger diameter than SMF and conventional splicing collapses hollow core

Initial un-normalized CO₂ measurements made using one segment of the inline fiber sensor.

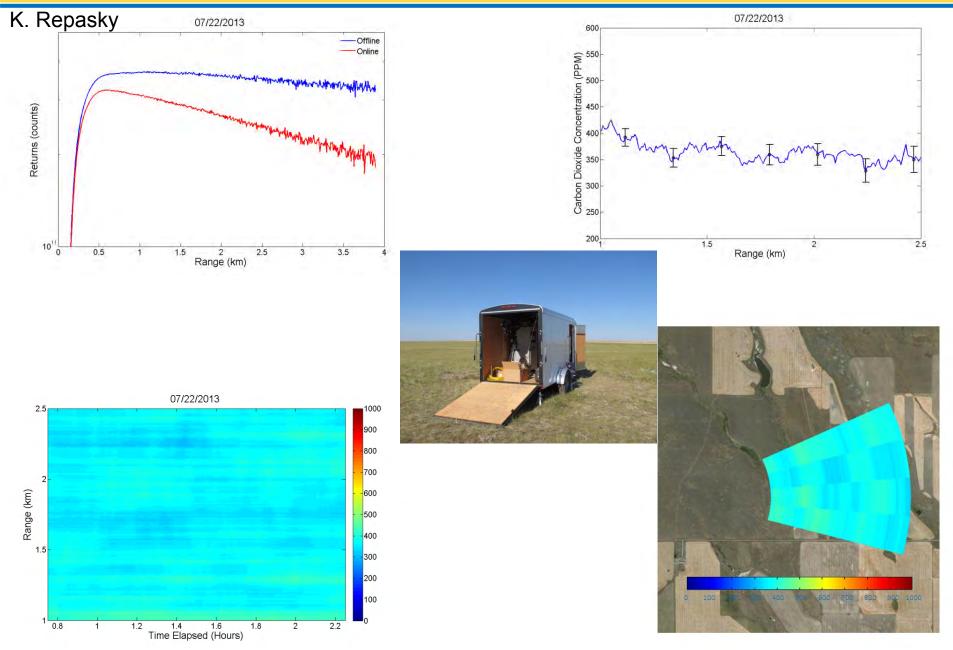


The PBG fiber allows interaction of the laser light and CO_2 in the hollow core.





Differential Absorption Lidar



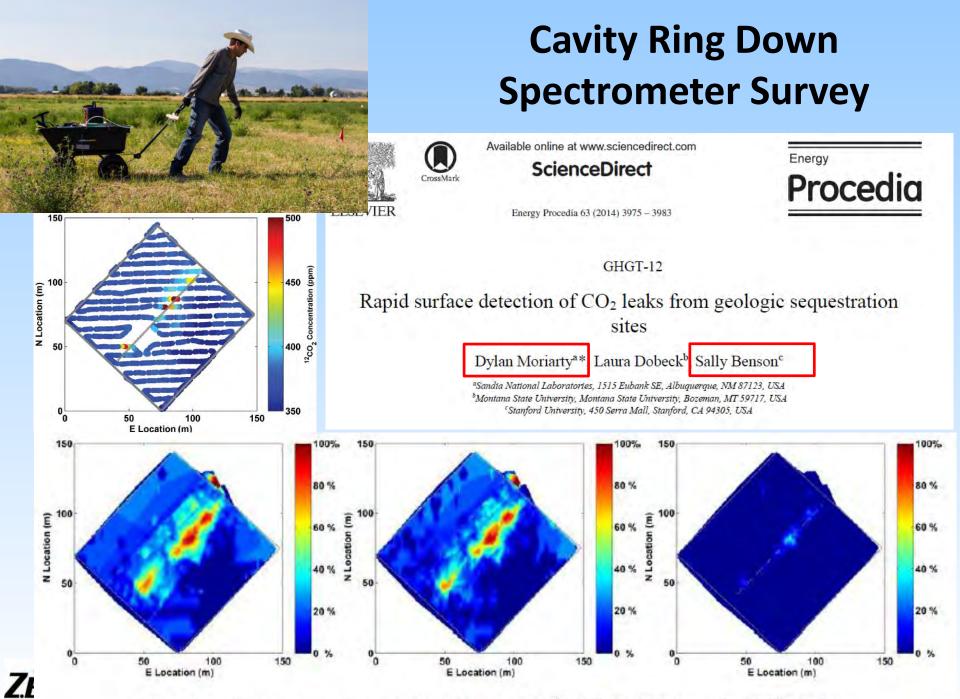
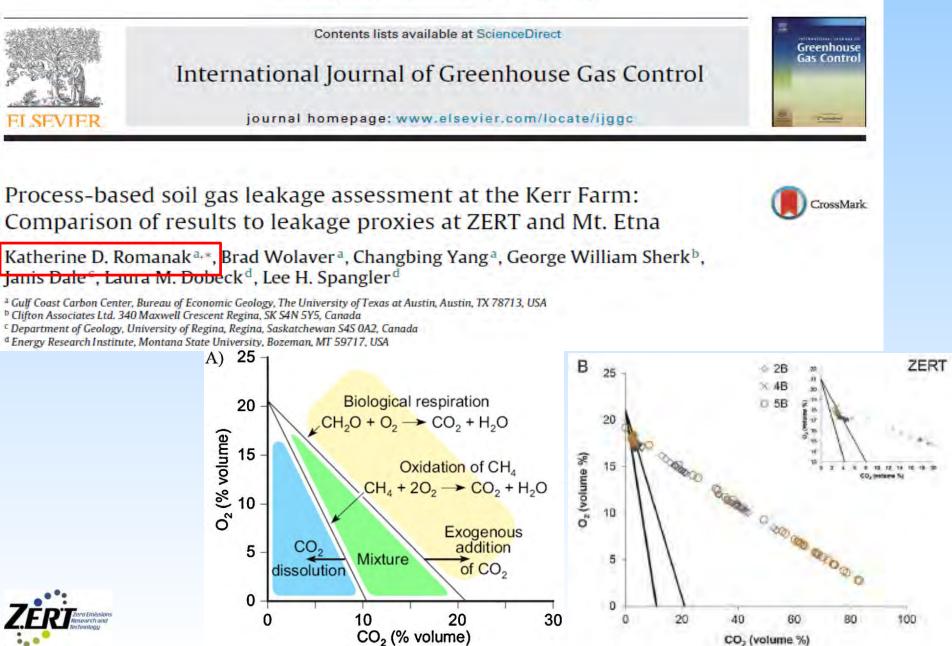


Fig. 6. Detection percentage using static threshold method for ¹²CO₂ (left), ¹³CO₂ (center), and δ¹³C (right).

Process Based Method

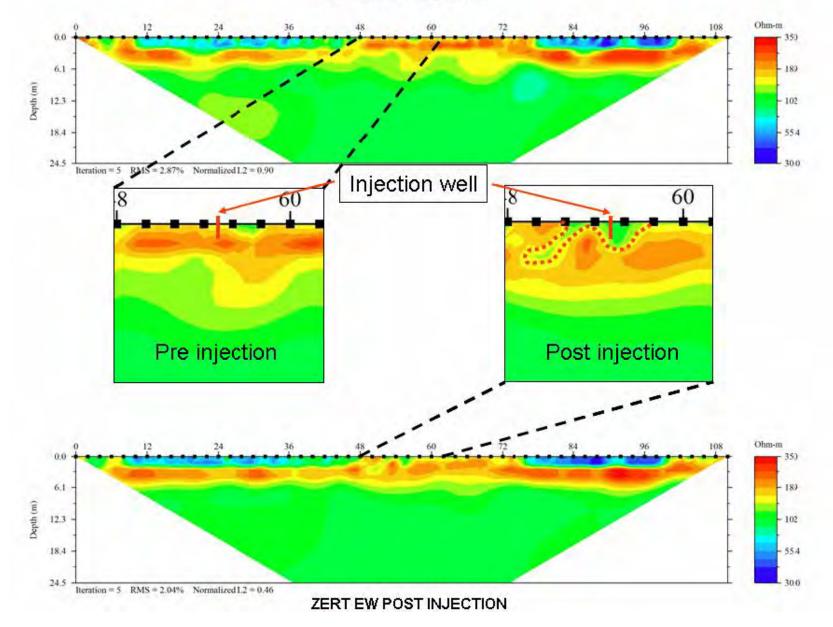
International Journal of Greenhouse Gas Control 30 (2014) 42-57



Resistivity

ZERT EW PREINJECTION





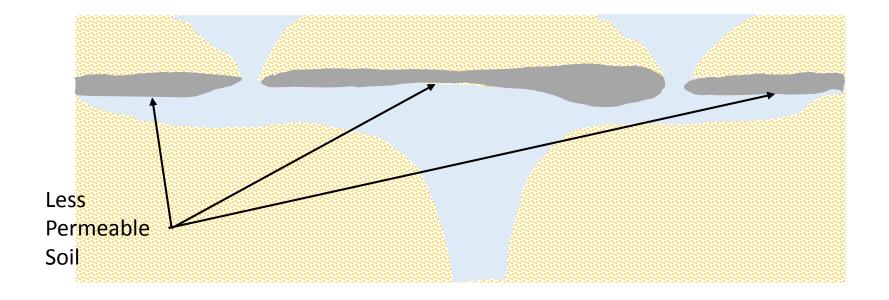
What We Have Learned

- Many near surface methods are quantitative <u>but</u>
 - Diurnal, seasonal, annual variations in ecosystem background flux affect detection limits
 - Appropriate area integrated, mass balance is a challenge
- Nearly all methods could detect 0.15 tonnes/day release at ZERT Atmospheric signals drop rapidly away from the ground surface
- Isotopes & tracers have lower detection limits than straight CO_2 flux or concentration
- Scaling, 6 tonnes per day would be detectable over an area 40 times as large
- Surface expression was "patchy" 6 areas of ~5m radius
- Natural analogs also seem to have "patchy" surface expression
- <u>By comparing multiple controlled release sites we see that</u> <u>different ecosystems respond somewhat differently</u>



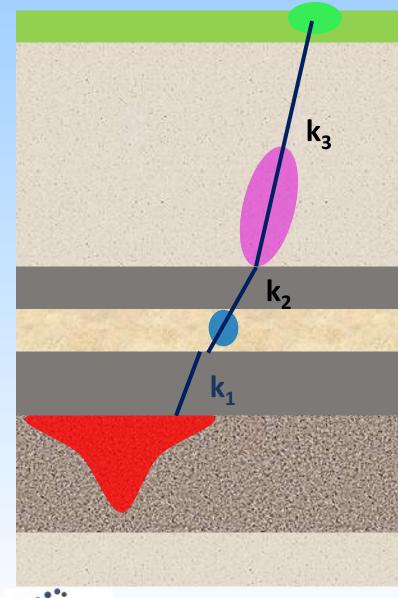


Why is the Surface Expression "Patchy"



If the horizontal permeability is significantly less than the vertical permeability, CO_2 will spread laterally until it hits a lower permeability vertical path. It can then desiccate that path creating a "chimney"

If CO₂ Escapes the Reservoir

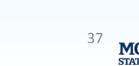


Many processes could prevent it from reaching the surface including

- 1. Trapping under a secondary seal
- 2. Geochemical conversion of CO_2
- 3. Dissolution

If it does reach the surface:

- 1. The surface flux will not necessarily be the same as the flux leaving the reservoir
- 2. The surface expression could be some distance from the storage reservoir



Monitoring – A Multi-Step Process

- Initial Detection (Finding anomalies in a large area.)
 - Wide Area Hyperspectral Imaging, Atmospheric tomography
 - Moderate Area Lidar, Fiber sensors, Resistivity
- <u>Confirmation</u> (Is anomaly due to elevated CO₂ flux?)
 - CO₂ flux and / or concentration measurements, water measurements
- <u>Attribution</u> (Is elevated flux due to leakage?)
 - Isotopic measurements
 - Process based measurements (relationships between multiple gases, Romanack)
- <u>Mapping and Quantification</u>
 - Flux chamber
 - Concentration measurements in a survey mode
- <u>Impact Measurement</u> Dependent on the receptor





What Is the Monitoring Purpose?

- Climate change mitigation?
 - 1% over 1000 yrs climate models?
- Retention in the reservoir?
 - Subsurface techniques typically do not measure properties directly proportional to concentration / quantity
- Overall storage security?
- HSE, Resource protection (USDW)?
 - Measure to ensure levels are below impact levels
- Public assurance?
- Verification and accounting?
 - Mass flow meters only accurate to $\sim 1\%$

If this is the primary focus, this could reduce need for wide – area monitoring.



How We Have Learned

Natural Analogs

- Mammoth Mountain
- Laacher See
- Latera
- Soda Springs, ID
- Crystal Geyser, UT
- More

How analogous is the analog?

Flow through significant overburden

Fluxes may be much higher than leaky engineered system

Controlled Releases

- ASGARD (Nottingham)
- ZERT (Montana State)
- Australia
- Norway
- More

Source term known Ability to establish detection limits Relatively little overburden







Acknowledgement

U.S. Department of Energy

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