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Validation of Models Simulating Capillary and Dissolution Trapping During Injection and Post-Injection of CO2 in Heterogeneous Geological Formations Using Data from Intermediate Scale Test Systems

(DE-FE0004630)

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Validation of Models Simulating Capillary and Dissolution Trapping During Injection and Post-Injection of CO_2 in Heterogeneous Geological Formations Using Data from Intermediate Scale Test Systems

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Presentation Outline

- **Project Overview** - Goals & Objectives
  - Benefits of technology
  - Project status
- **Key questions and knowledge gaps**
  - Successful storage
  - Role of models in design
  - Conceptualization and key questions
- **Objectives and tasks**
- **Multi-scale physical and numerical modeling approach**
  - Experimental methods
- **Technical progress and results**
  - Capillary trapping
  - Dissolution trapping
- **Findings**
- **Future Plans**
- **Appendix**

Benefits of Technology to the Program

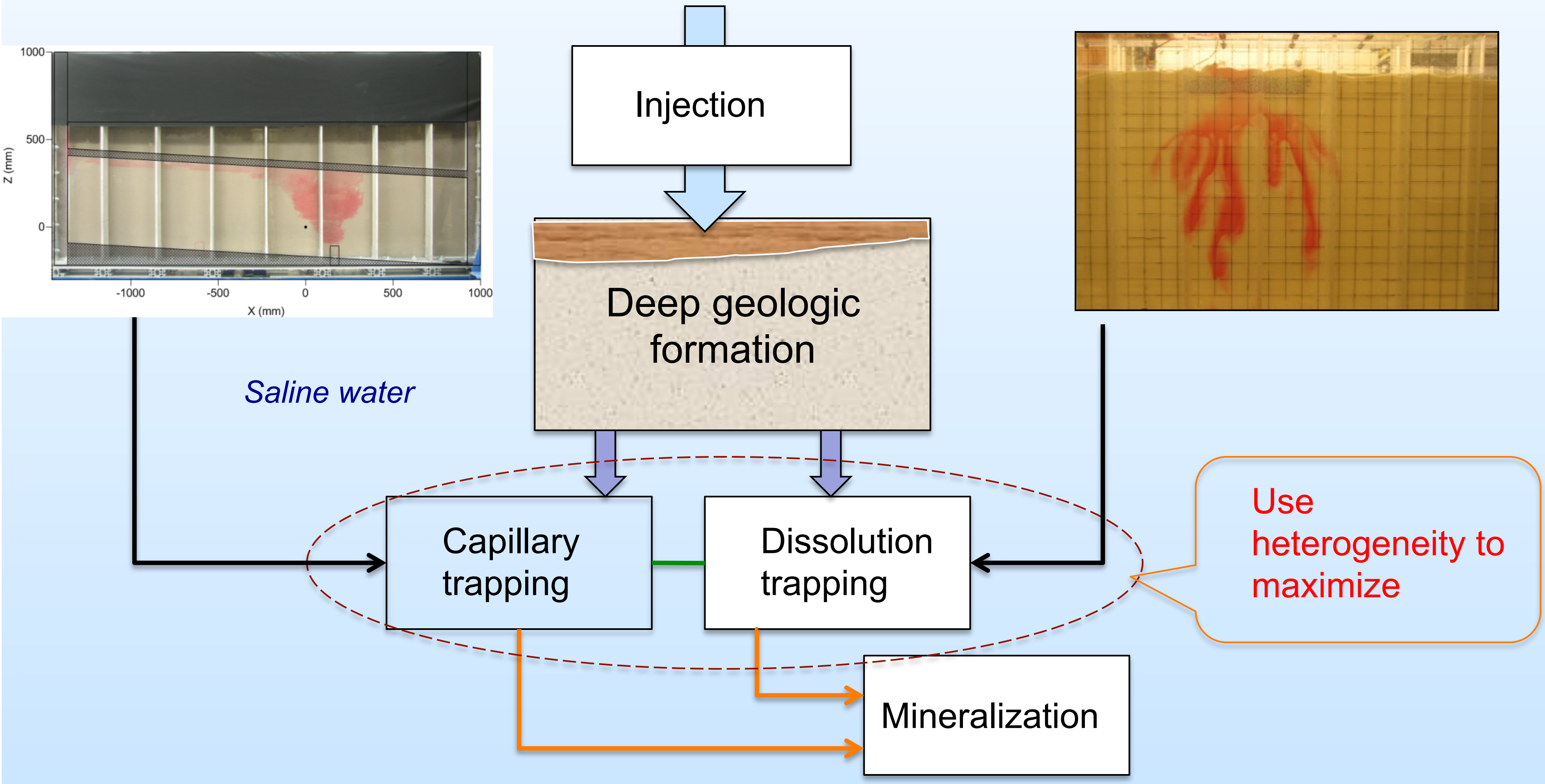
- Improve/develop and validate models by using data generated in intermediate-scale laboratory test systems (~1 - 5m length) simulating capillary and dissolution trapping under various heterogeneous conditions.
- Design injection strategies, estimate storage capacities and efficiency for field-scale geological systems by using the improved numerical tools.
- The findings will meet objectives of Program research to develop technologies to cost-effectively and safely store and monitor CO2 in geologic formations and to ensure storage permanence.
- Developed approach and technologies in this project specifically contribute to the Carbon Storage Program's effort of supporting industries' ability to predict geologic storage capacity to within +/- 30 percent.

Project Overview:
Goals and Objectives

- Objectives
  - Investigate how the trapping mechanisms are affected by formation heterogeneity with the ultimate goal of contributing towards improving numerical tools leading to up-scaling methods to design injection strategies, estimate storage capacities and efficiency, and conduct performance assessment for stable storage.
- Goals
  - The mechanisms of capillary and dissolution trapping that are affected by heterogeneity will be investigated using intermediate scale testing in porous media tanks.
  - The generated data will be used to improve the conceptual understanding and develop and validate models that will allow more accurate prediction of CO2 fate and transport in deep geologic saline formations.

Successful Storage

The goal of successful storage is to create stable conditions where the CO_2 becomes immobilized through entrapment, dissolution, and mineralization.



Research Questions

Capillary Trapping

- ❑ How do heterogeneities and connectivity (spatial continuity of different permeability zones) affect entrapment efficiency of $scCO_2$ in deep geological formations?
- ❑ How different injection schemes can be adopted to improve storage efficiency via capillary trapping?
- ❑ How well the existing continuum-based models and the constitutive models capture multiphase (water/$scCO_2$) flow behavior in deep formations?

Dissolution Trapping

- ❑ Under what geologic conditions convective mixing is important?
- ❑ What are the effects of heterogeneity on dissolution and density-driven fingers?
- ❑ What is the contribution of low permeability zones to the stable trapping of dissolved CO_2?
- ❑ Can mass loading in the CO_2 capillary entrapment zone be represented using effective mass transfer rate coefficient?

Project Overview- Scope of Work

- Generate a comprehensive data set in intermediate scale test tanks simulating multiphase flow to investigate how effective capillary trapping is affected by the texture transitions and variability in heterogeneous field formations.

- Generate a comprehensive data set in intermediate scale test tanks simulating dissolution of partially miscible fluids to investigate how effective dissolution trapping is affected by heterogeneity-driven preferential flow and cross-intra-layer mixing.

- Modeling efforts that include various scenario simulations to evaluate whether the existing modeling codes can accurately capture processes observed in the test tanks. This effort will lead to develop up-scaling methods for larger-scale applications.

Project Overview: Scope of Work

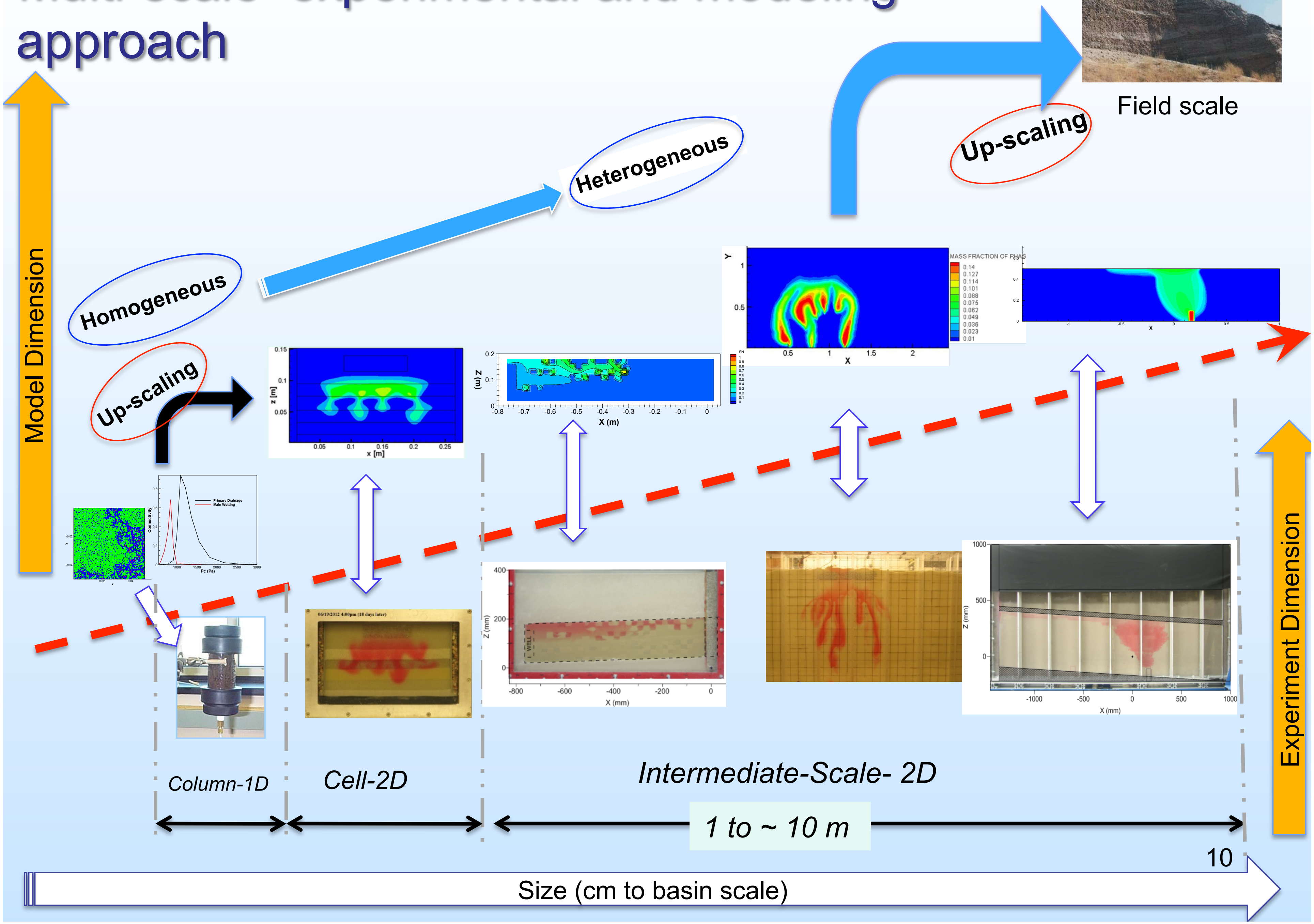
Why has the Scope of Work been defined as it is?

- Even though trapping at the core-scale is reasonably well understood and empirically modeled for relatively homogenous systems, critical knowledge gaps exist on how these processes manifest themselves under conditions of ubiquitous field heterogeneities to estimate or predict effective trapping capacities of field systems.

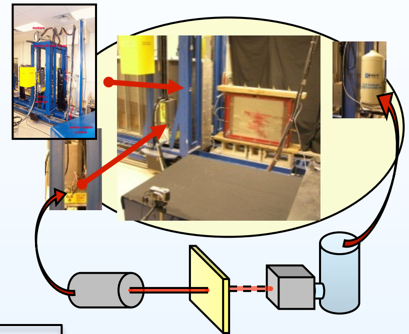
- Comprehensive understanding of the CO_2 storage and entrapment problem is only possible through multistage analysis comprising of experimental studies under highly controlled conditions and modeling.

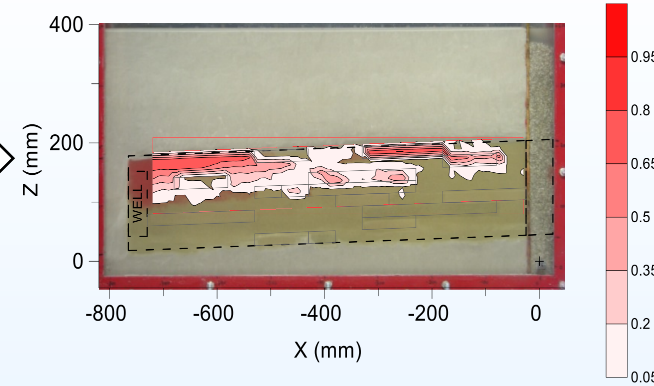
- To our knowledge, none of the existing modeling tools have been validated or tested for their ability to accurately capture the CO_2-brine-water flow patterns and entrapment mechanisms in porous media, specifically under heterogeneous conditions.

Multi-scale experimental and modeling approach

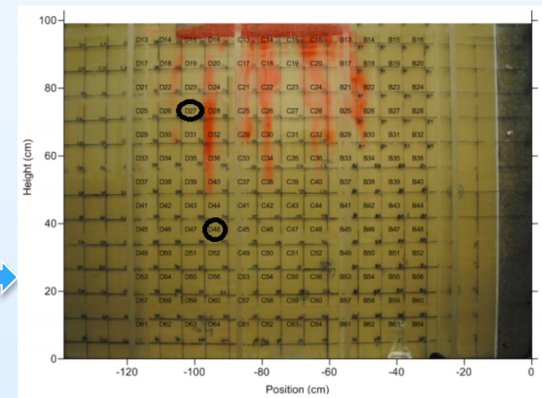


Experimental Methods



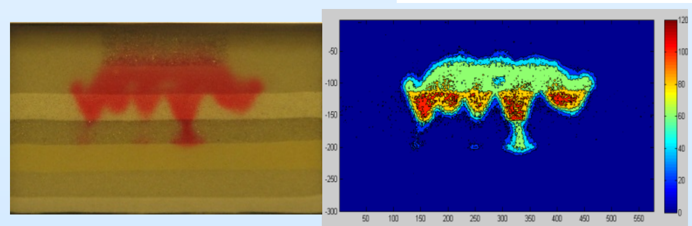


- **Automated transient and spatially distributed saturations using x-ray attenuation**



- **Aqueous sampling to determine dissolved plume concentrations.**

- **Image processing to determine the dissolved plume concentrations.**



- **Measurement of multiphase model parameters (capillary pressure-saturation-relative permeability relationships)**

Capillary trapping

- ❑How do heterogeneities and connectivity (spatial continuity of different permeability zones) affect entrapment efficiency of $scCO_2$ in deep geological formations?
- ❑How different injection schemes can be adopted to improve storage efficiency via capillary trapping?

Progress and results

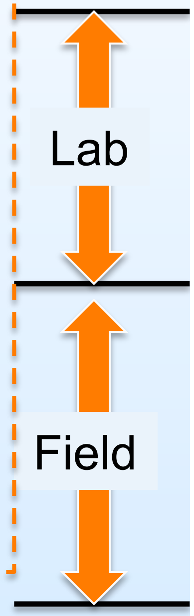
- ✓ Surrogate fluid selection
- ✓ Small tank experiments to evaluate effects of heterogeneity
- ✓ Trapping during forced imbibition
- ✓ Large tank experiments – Homogenous
  - Heterogeneous
- ✓ Modeling and simulations

Surrogates for Trapping of $ScCO_2$ in Brine

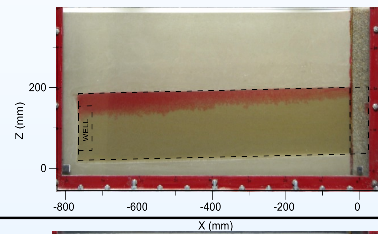
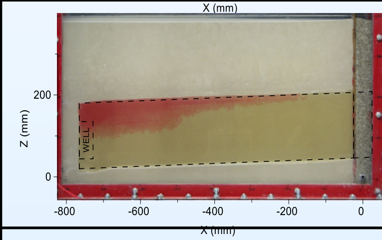
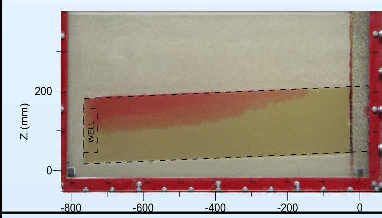
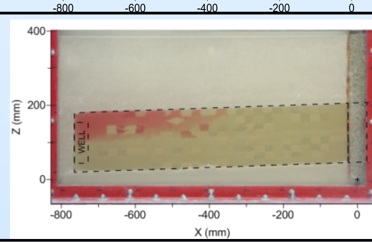
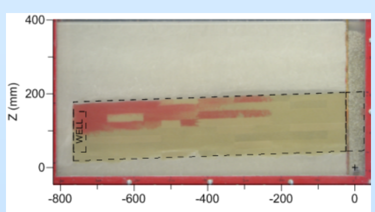
- Laboratory investigation of $scCO_2$ migration without high pressure in deep formations can be conducted using analogous fluids having similar density and viscosity contrasts as $scCO_2$ – brine phases under storage conditions

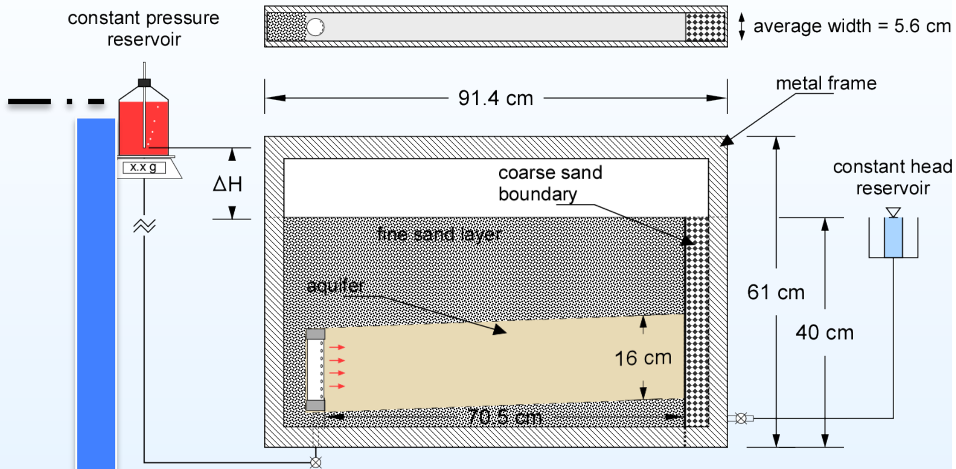
phase	ρ ($kg\ m^{-3}$)	μ (mPa· s)	μ_{nw}/μ_w	ρ_{nw}/ρ_w	IFT ($mN\ m^{-1}$)
Soltrol 220	860	4.9	0.072	0.71	15
Glycerol-water	1210	68			
$scCO_2$	266-733[a] (760[c])	0.023-0.0611[a] (0.06[c])	0.026-0.20[a] (0.075[c])	0.22-0.75[a] (0.745[c])	19.8[b]
Brine	945-1230[a] (1020[c])	0.195-1.58[a] (0.8[c])			

[a] estimates from Nordbotten et al. (2005), T = 35-155°C, P = 10.5-31.5 MPa
[b] measurement from Bennion and Bachu (2006), T = 43°C, P = 20 MPa, brine salinity = 2.7% wt.
[c] estimates from Singh et al. (2010) for Sleipner field



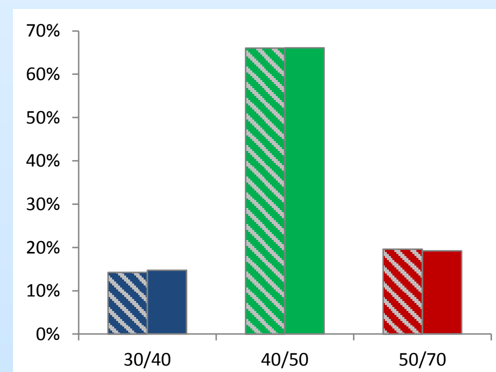
Effect of heterogeneity on trapping - 3'x2' tank experiments

#1	Homogeneous (#30/40 Accusand)	
#2	Homogeneous (#40/50 Accusand)	
#3	Homogeneous (#50/70 Accusand)	
#4	Heterogeneous (#30/40, #40/50, #50/70)	
#5	Heterogeneous (#30/40, #40/50, #50/70)	

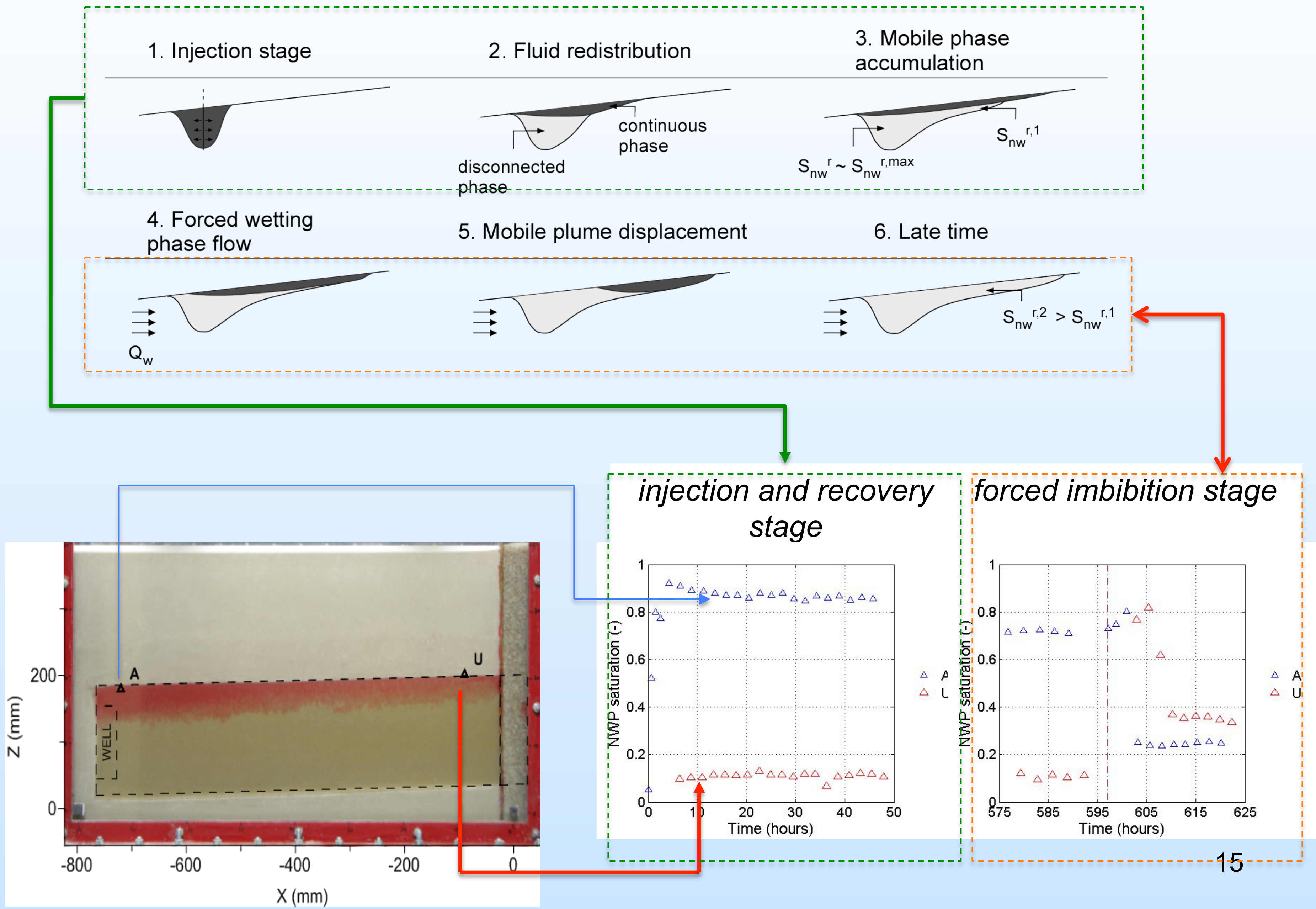


Corse to fine sand

Low to high correlation length

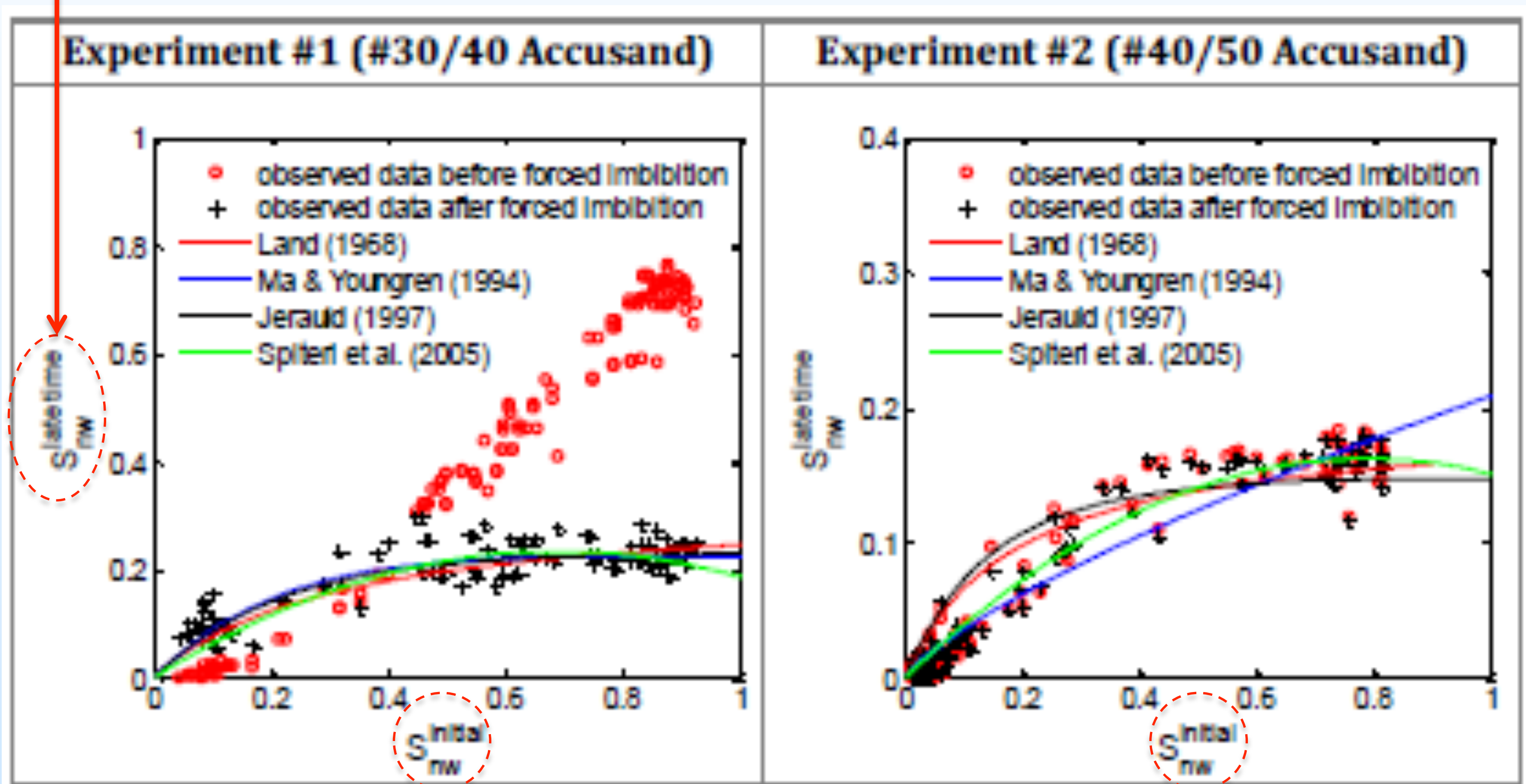


$ScCO_2$ saturation evolution during forced imbibition



Trapping in homogeneous formations after forced imbibition

Trapped saturation after flushing $S_{nw}^{late\ timel}$

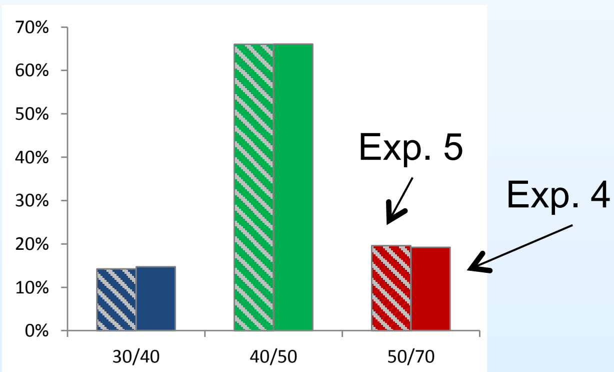


Trapped saturation before flushing $S_{nw}^{initial}$

from Trevisan et al. (2014)

Effect of forced imbibition/chase brine

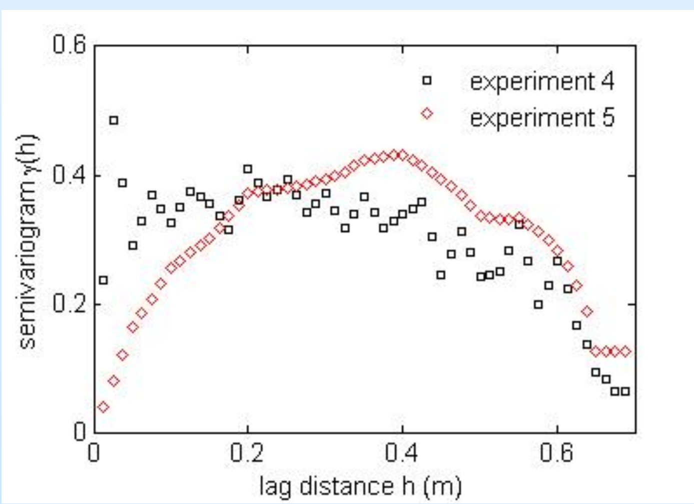
Same sand proportions for both heterogeneous experiments:
14% of #30/40 (coarse sand)
66% of #40/50 (intermediate sand)
20% of #50/70 (fine sand)

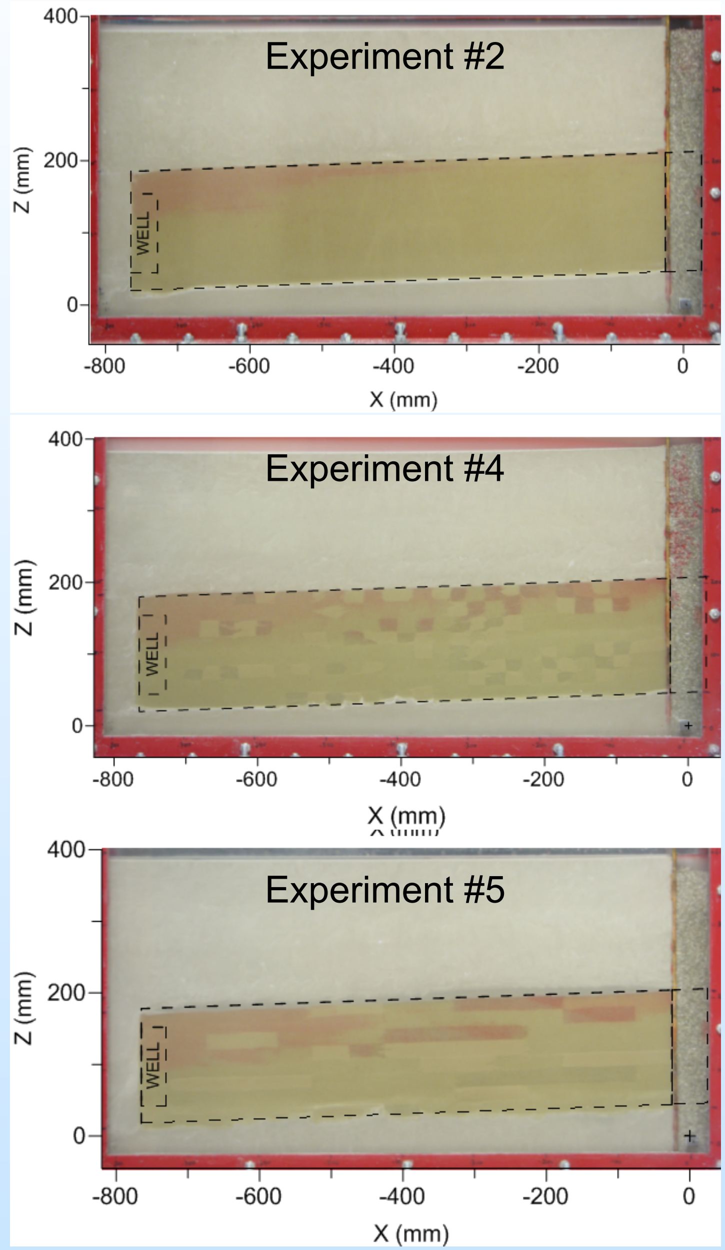


After 48 hours of forced imbibition

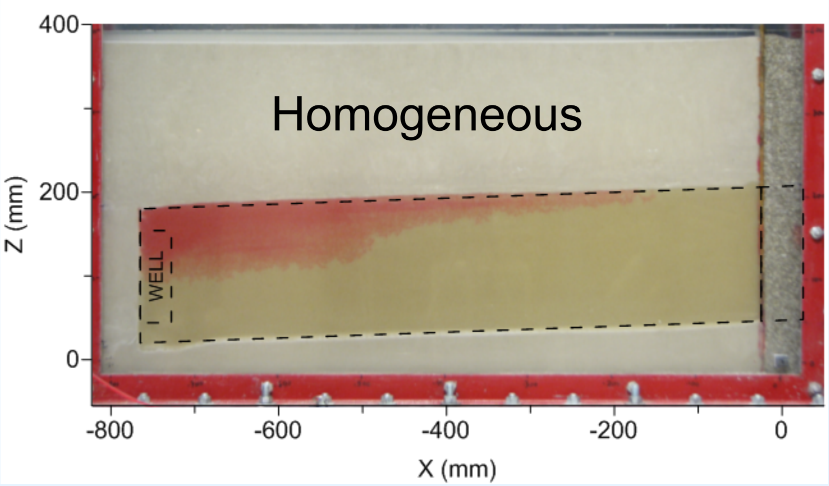
After 5.5 hours of injection

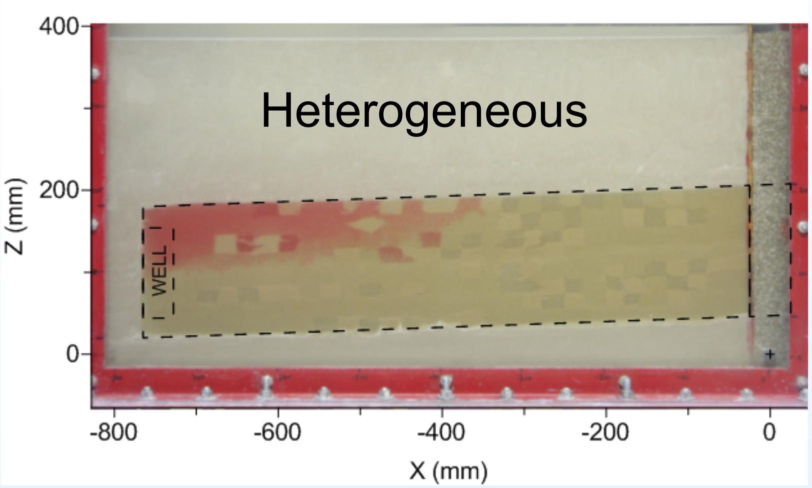
Experimental variograms: increased nugget effect for experiment #4

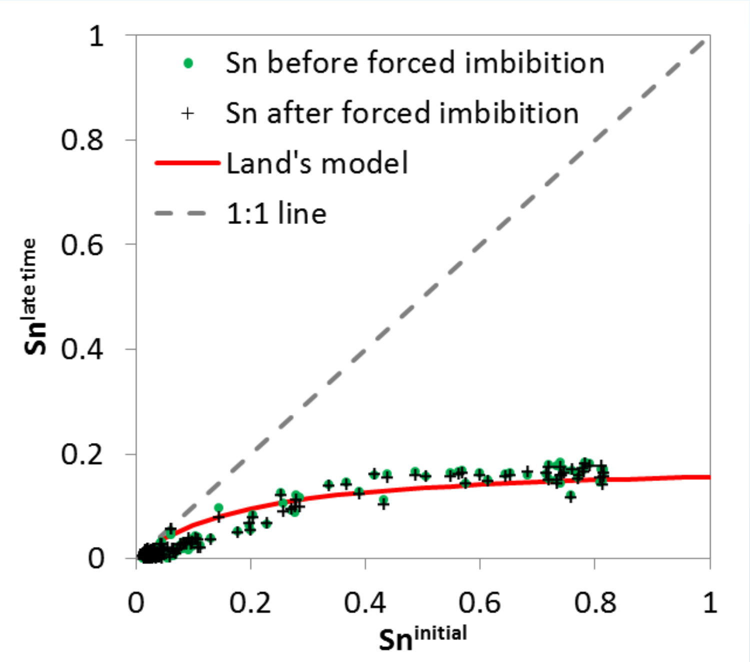


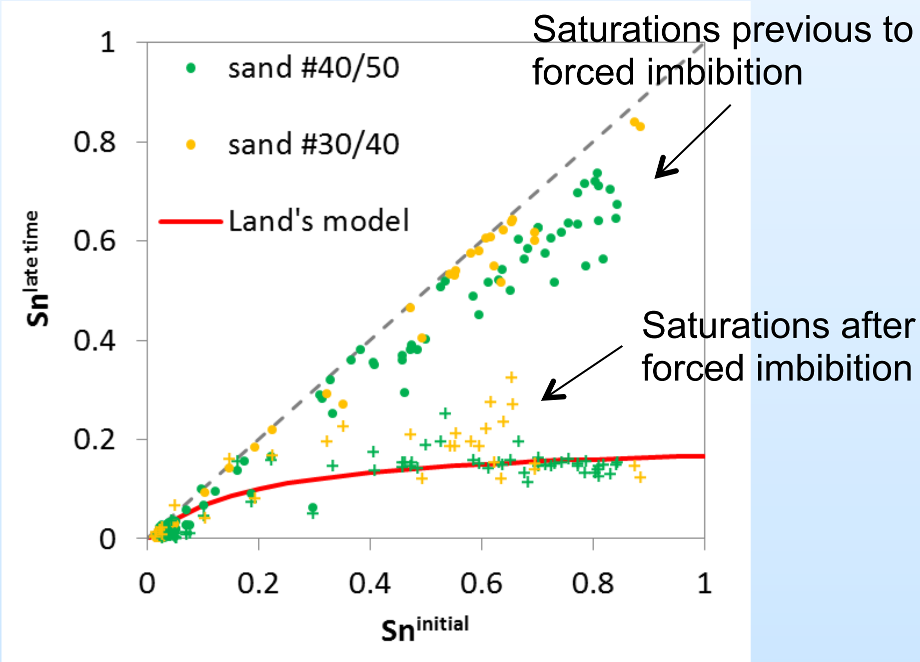


Capillary barriers enhancing $ScCO_2$ trapping



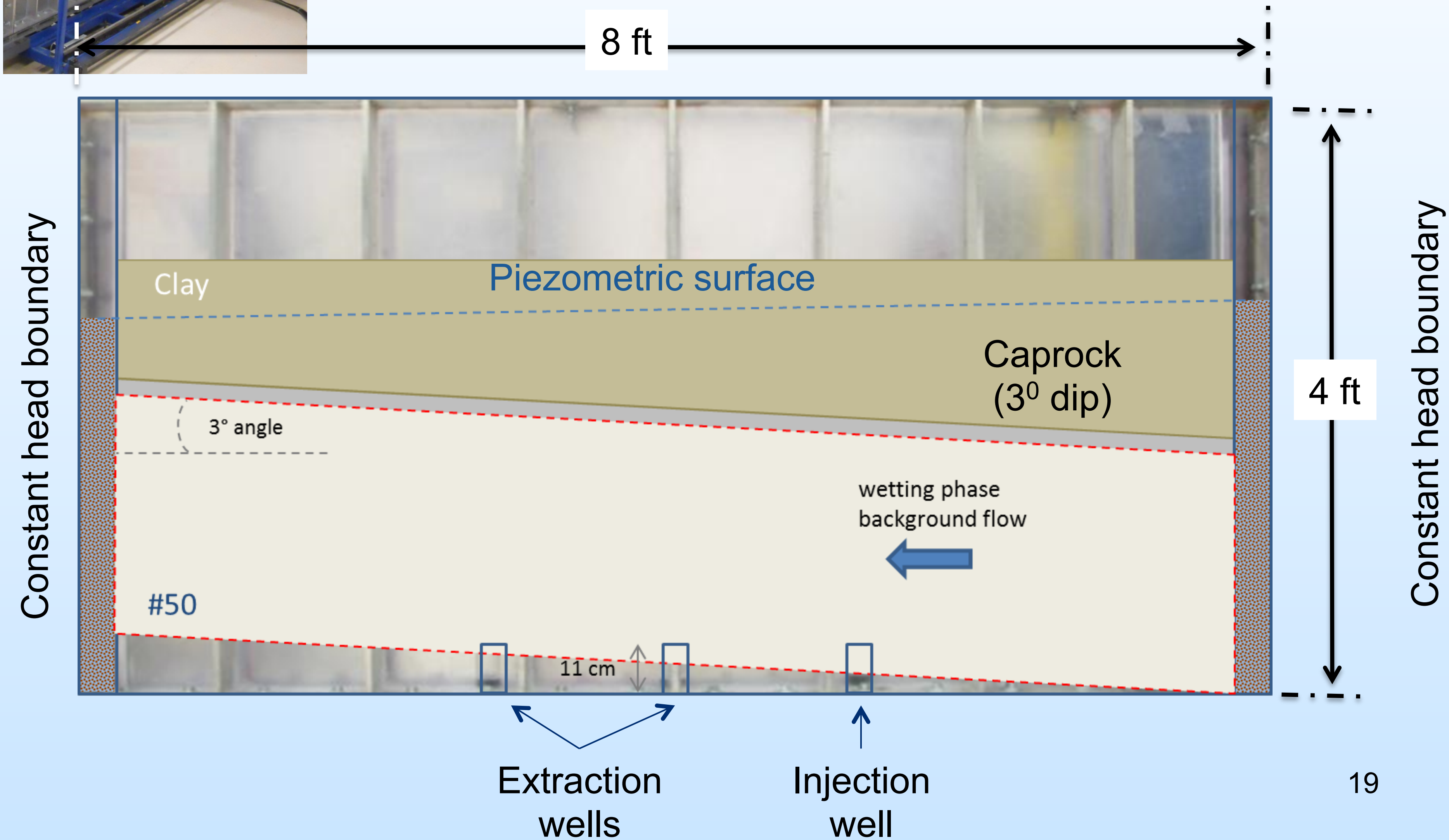




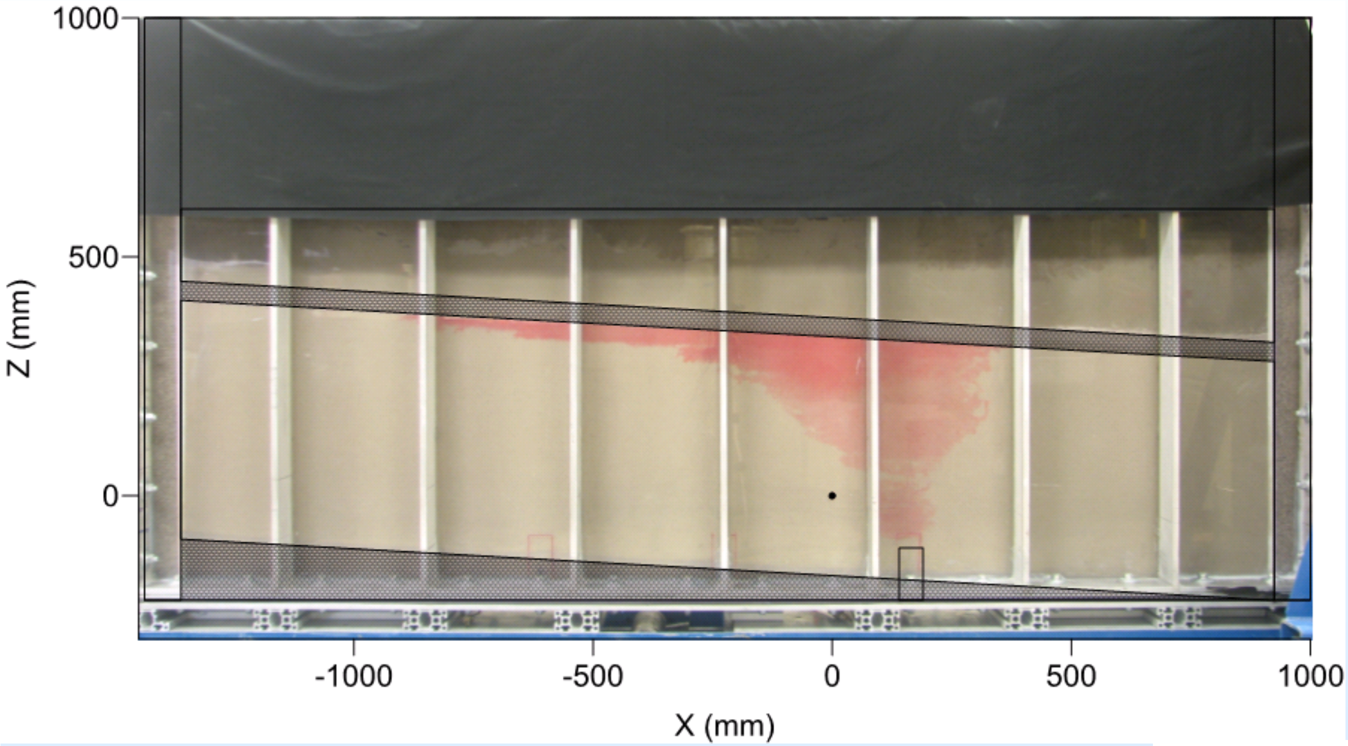


Up-scaling to large tanks

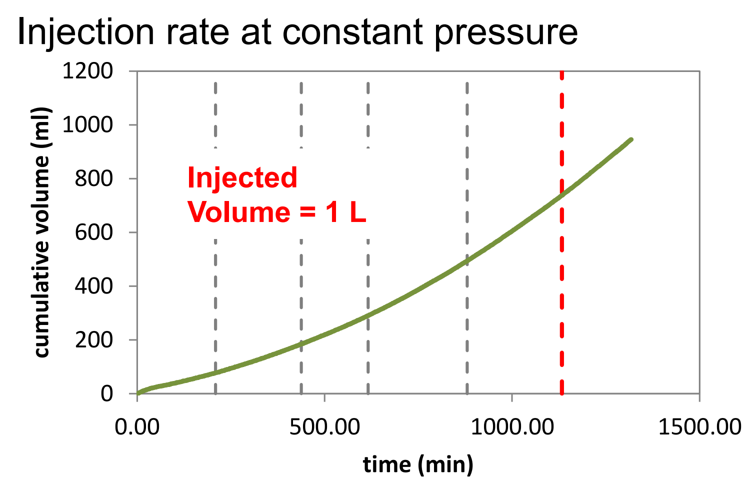
- Homogeneous packing with # 50 sand
- Use of x-ray attenuation to accurately map NWP saturations
- Re-injection after full immobilization



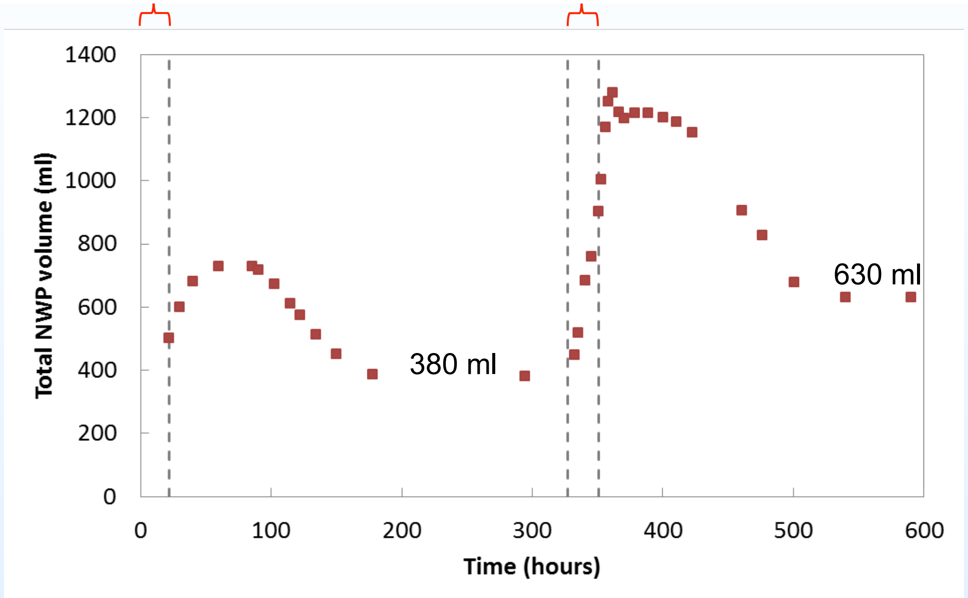
t = [illegible] hours



- ✓ Visible effect of wetting phase flow gradient (right to left)
- ✓ Injection rate increases with time under constant injection pressure

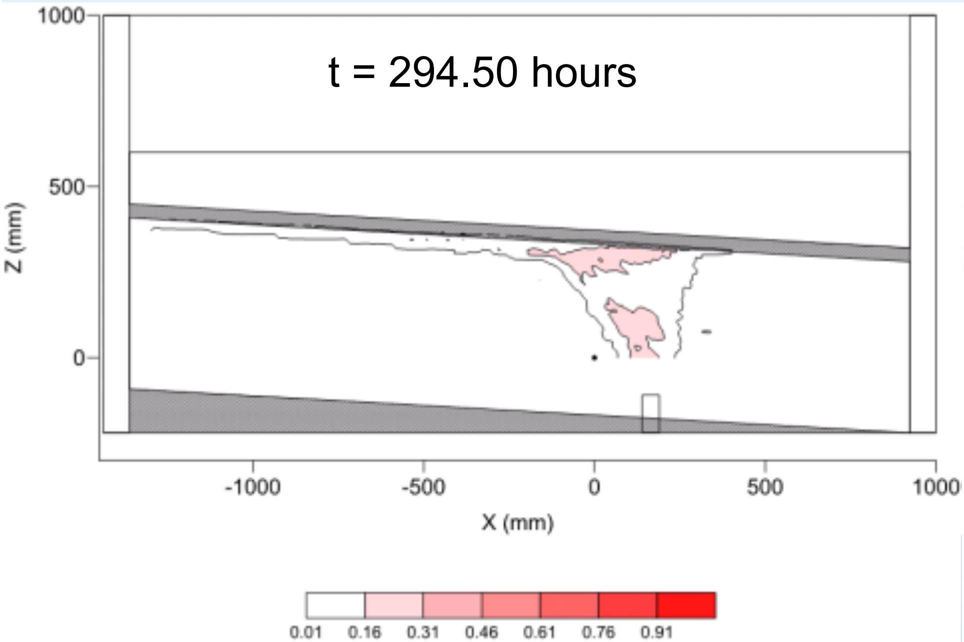


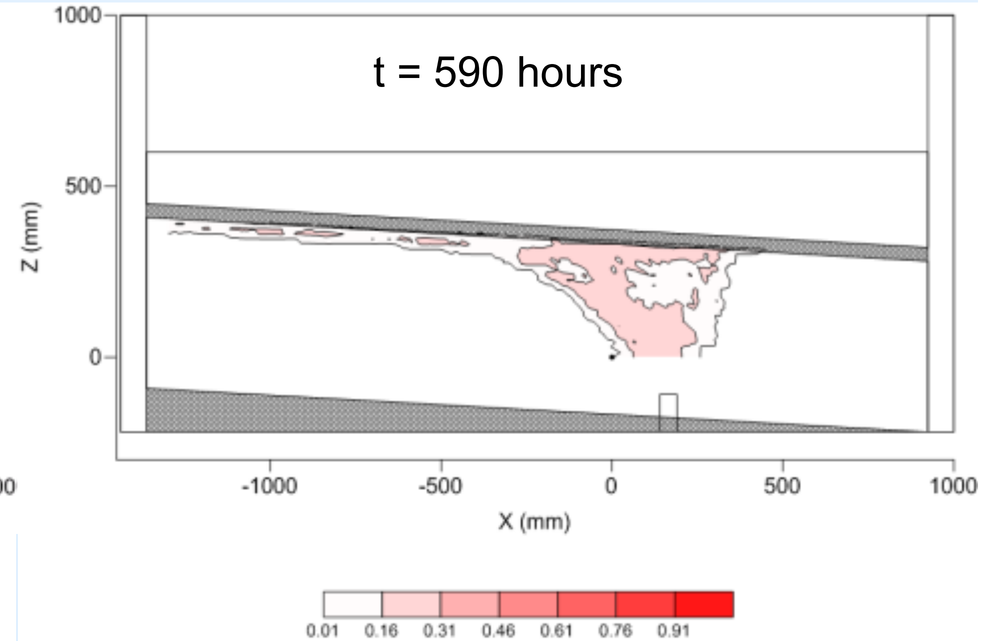
Total volume of trapped NWP in the plume (0^{th} moment) plume (covered by scanning grid)



Residually trapped NWP **volume** is 60% higher after 2nd injection event

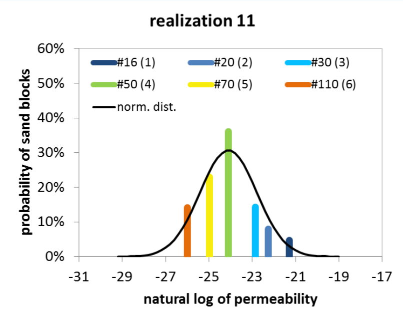
Residually trapped NWP **area** is 23% larger after 2nd injection event

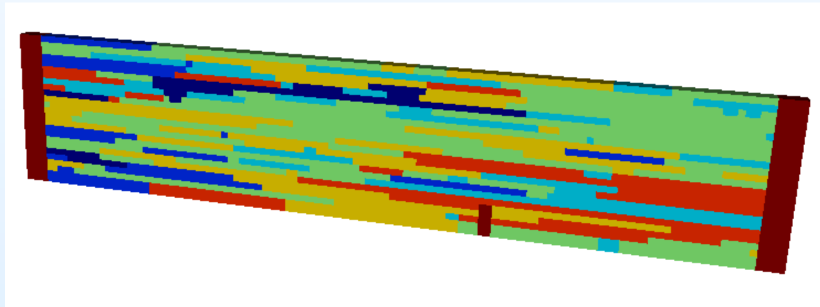


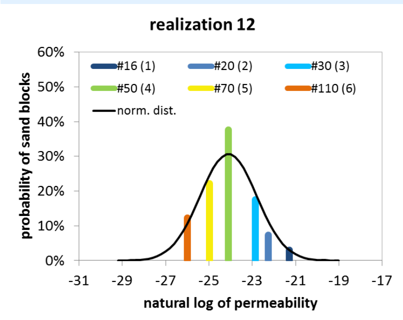


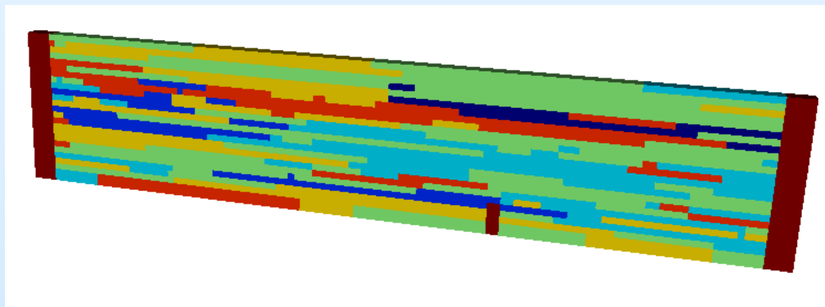
Design of a heterogeneous pack

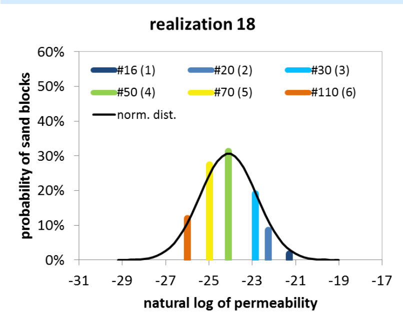
- A normal distribution of natural log of permeability (in m^2) with mean μ=-24.1 (#50 sand) and variance σ^2=1.3 is discretized in 6 categories
- Sequential Indicator Simulation is used to populate a 122x25 cells domain with average correlation length of 1 m (horizontal) and 4 cm (vertical)

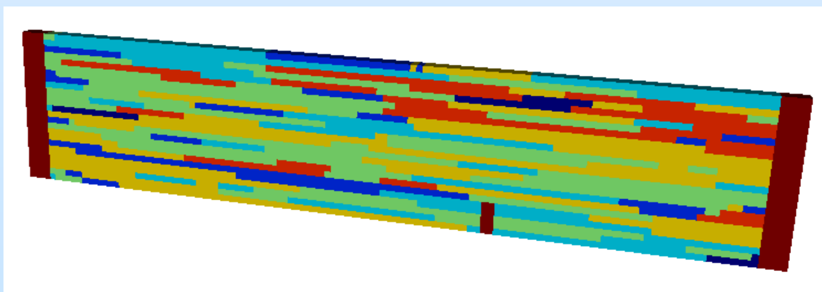




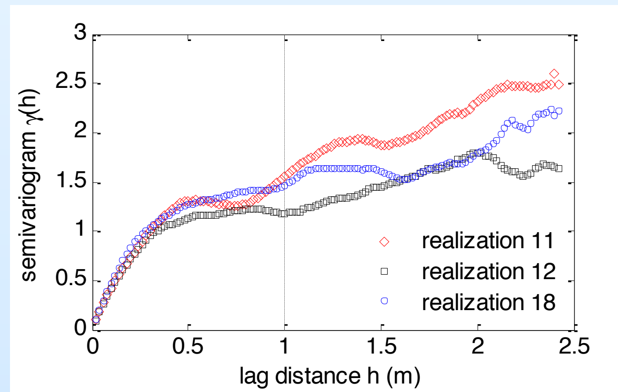








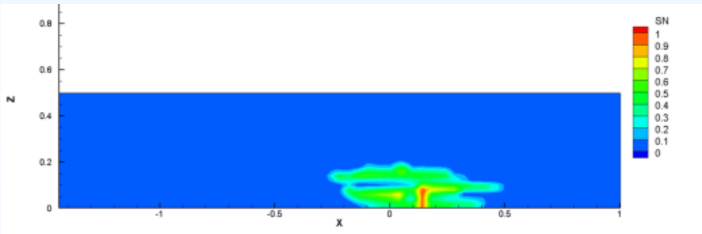
Semi-variogram



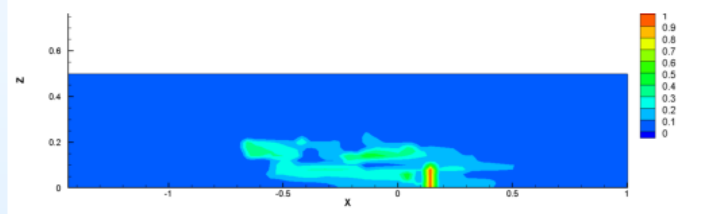
Numerical simulations

- ✓ Injection and fluid redistribution stages performed using the in-house code TPFLOW
- ✓ Selected configuration facilitated spreading of the plume and avoiding an early breakthrough into the constant head boundaries

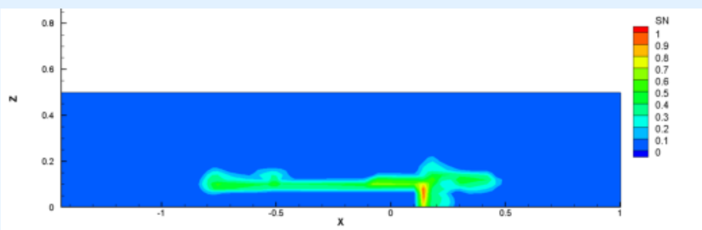
Scenario 11 – after 22 hours of injection



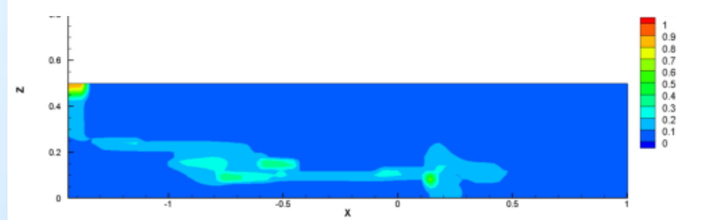
Scenario 11 – after 2 weeks of fluid redistribution



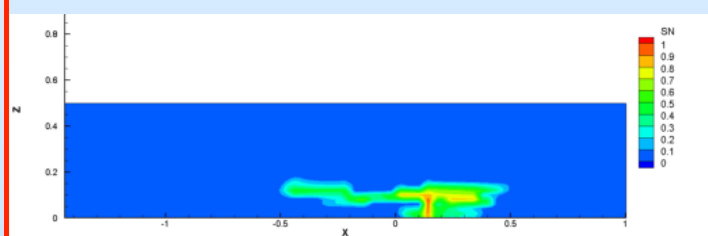
Scenario 12 – after 22 hours of injection



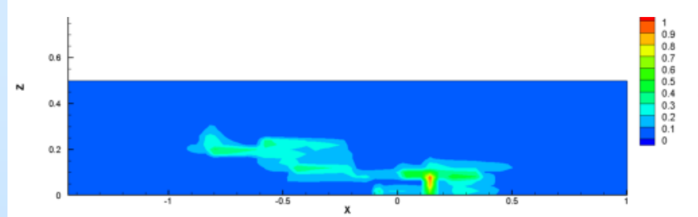
Scenario 12 – after 2 weeks of fluid redistribution



Scenario 18 – after 22 hours of injection



Scenario 18 – after 2 weeks of fluid redistribution



Selected Packing configuration

Mixing and dissolution trapping

- Under what geologic conditions convective mixing is important?
- What are the effects of heterogeneity on dissolution and density-driven fingers?
- What is the contribution of low permeability zones to the stable dissolution trapping?
- Can mass loading in the CO2 capillary entrapment zone be represented using an effective mass transfer rate coefficient?

Progress and results

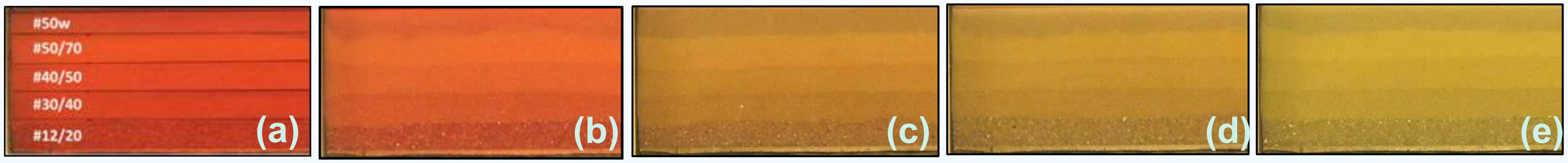
- ✓ Testing the new analog fluid combination
- ✓ Developing a numerical code for mixing
- ✓ Testing the sands in a small cell to be used in large tank experiments
- ✓ Testing the new injection system in small cell
- ✓ Investigating convective mixing and stable trapping in heterogeneous formations numerically and experimentally
- ✓ Performing the large tank experiments (low permeability zone storage)

Selection for Dissolution of $ScCO_2$ in Brine

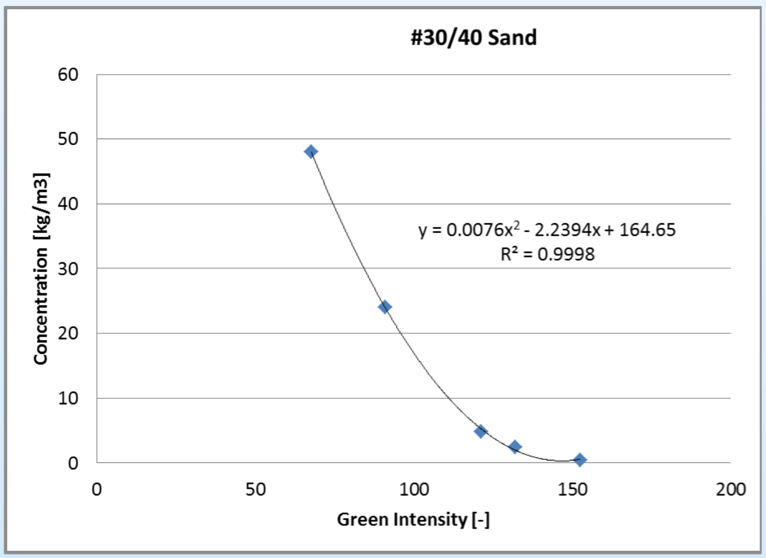
	scCO2-brine @ Typical Reservoir Conditions	Water in Propylene Glycol @ 20C, 1 atm
Viscosity Ratio $\frac{\mu_{nw}}{\mu_w}$	~ 0.05 - 0.2	~0.017
Density Ratio $\frac{\rho_{nw}}{\rho_w}$	~ 0.2 – 0.8	~0.9
Solubility	~3-5 %	miscible

- NaBr solution dyed with food dye and water is the second analog fluid combination.
- The solution was prepared to provides the same density contrast between formation brine and dissolved $scCO_2$ under storage temperature and pressure.

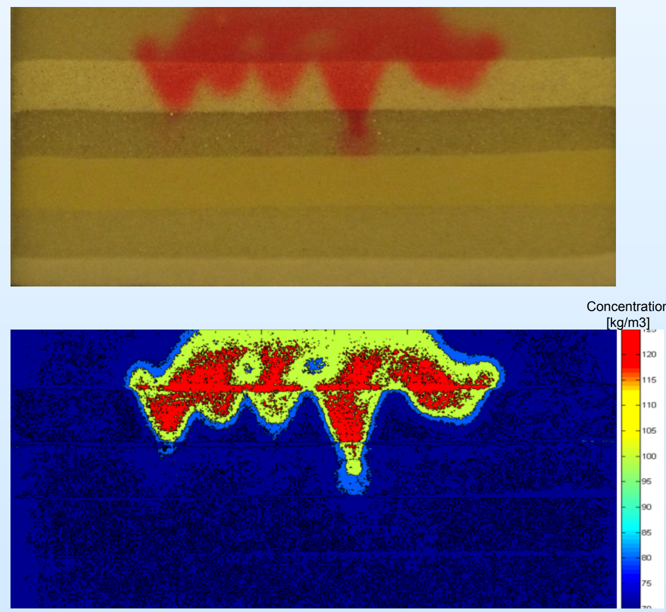
Image Processing



Five different sands having (a) 0.048 g/cm^3, (b) 0.024 g/cm^3, (c) 0.0048 g/cm^3, (d) 0.0012 g/cm^3 and (e) 0.00048 g/cm^3 concentrations



Concentration-Green intensity relationship for #30/40 sand.



Comparison of the experimental and image processing results for layered heterogeneous medium case (1 week results)

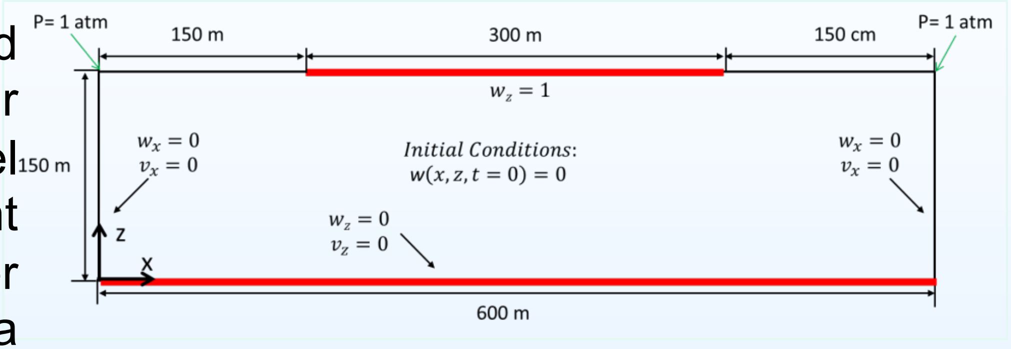
Numerical Model Development

- FVM-based, single-phase, two-component, and density and viscosity-dependent flow and transport model was developed to better understand the processes observed during convective mixing.

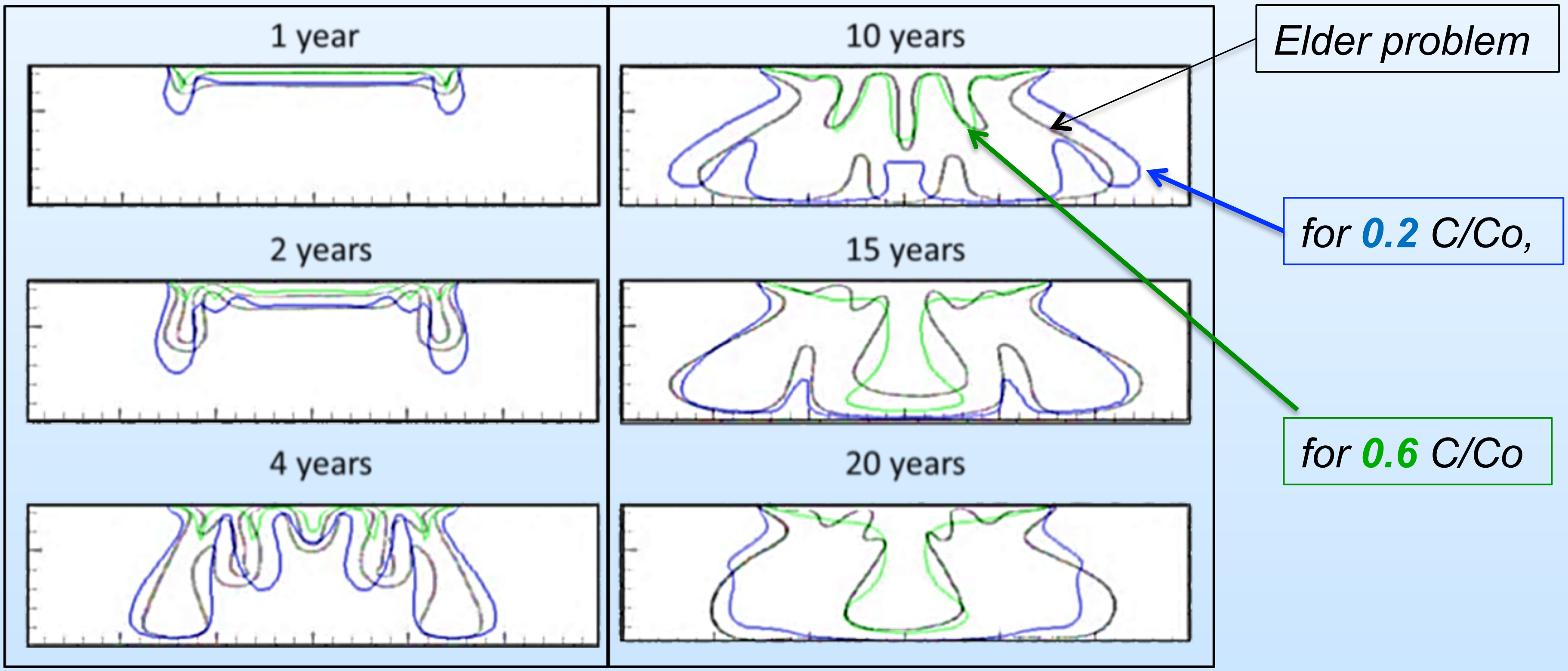
- To model the small tank experiments [*Agartan et al*., 2014a] accurately, the density and viscosity dependence on mass fraction of $scCO_2$ representative fluid (water) was included.

Model verification using the Elder Problem

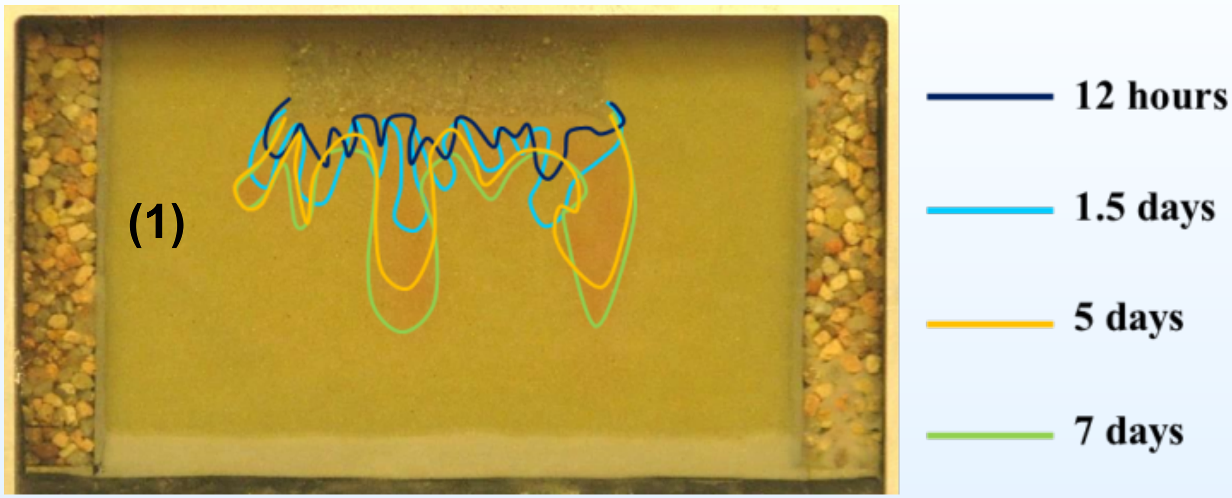
Voss and Souza [1987] re-formed *Elder* [1967] problem for laminar fluid flow in a box shaped model due to vertical temperature gradient as a variable density groundwater problem where fluid density is a function of salt concentration.

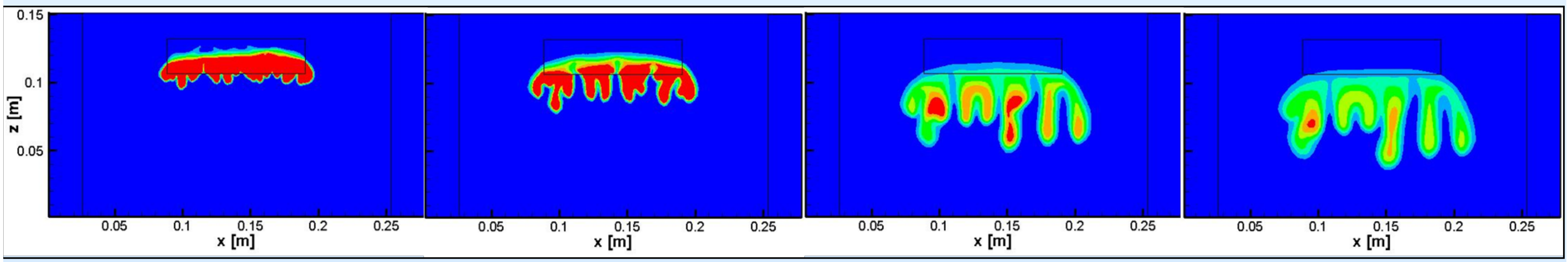


Initial and boundary conditions



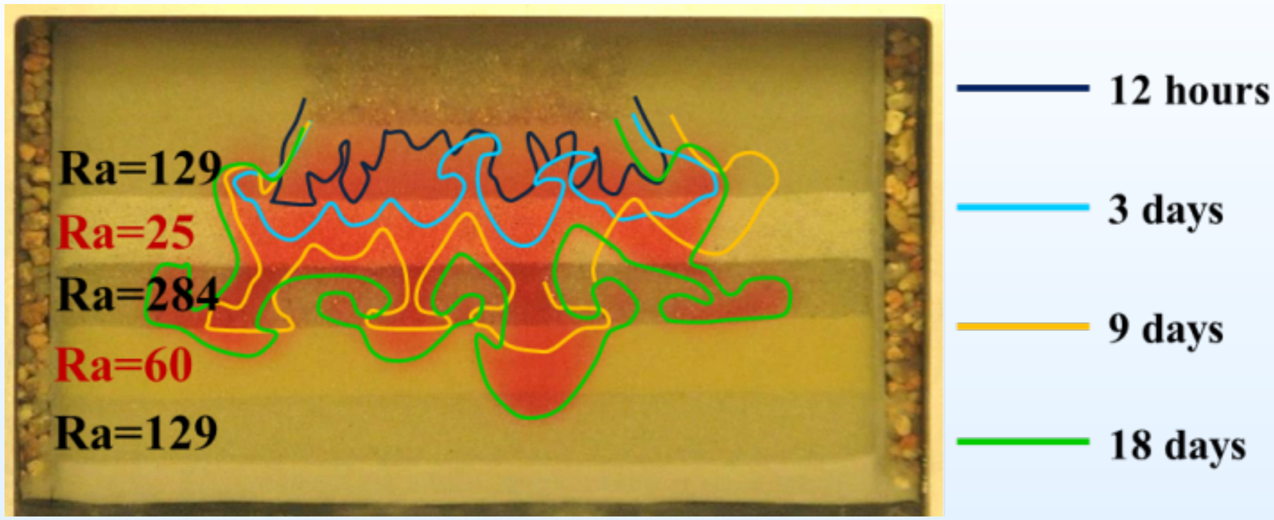
Modeling of the Small Tank (homogeneous)

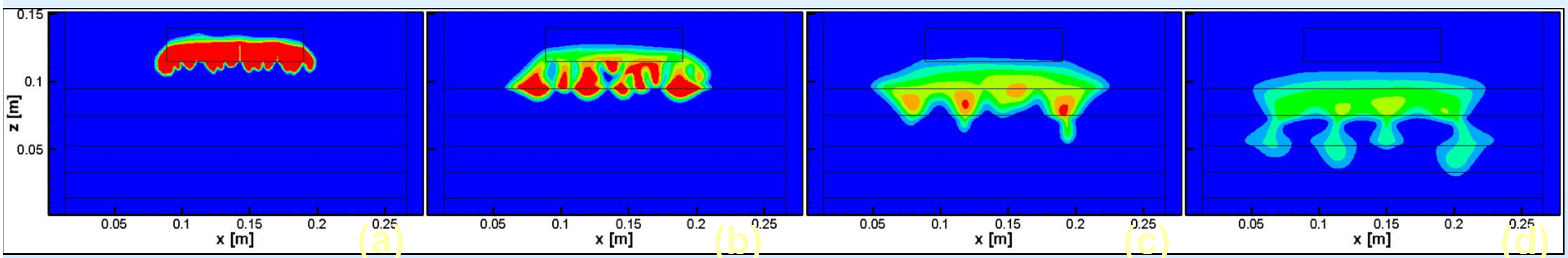




12-hour 1.5-day 5-day 7-day

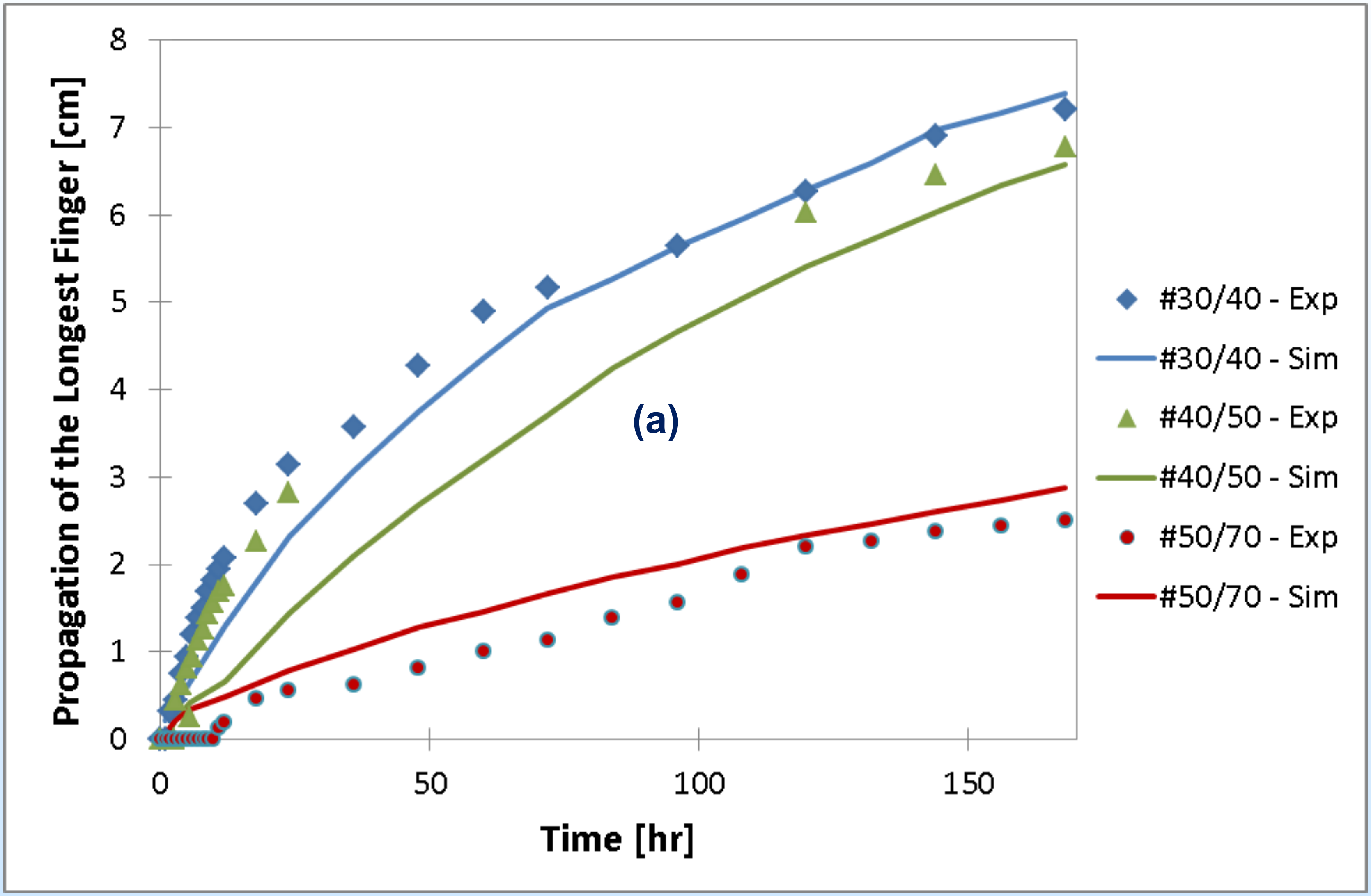
Modeling of the Small Tank (heterogeneous)



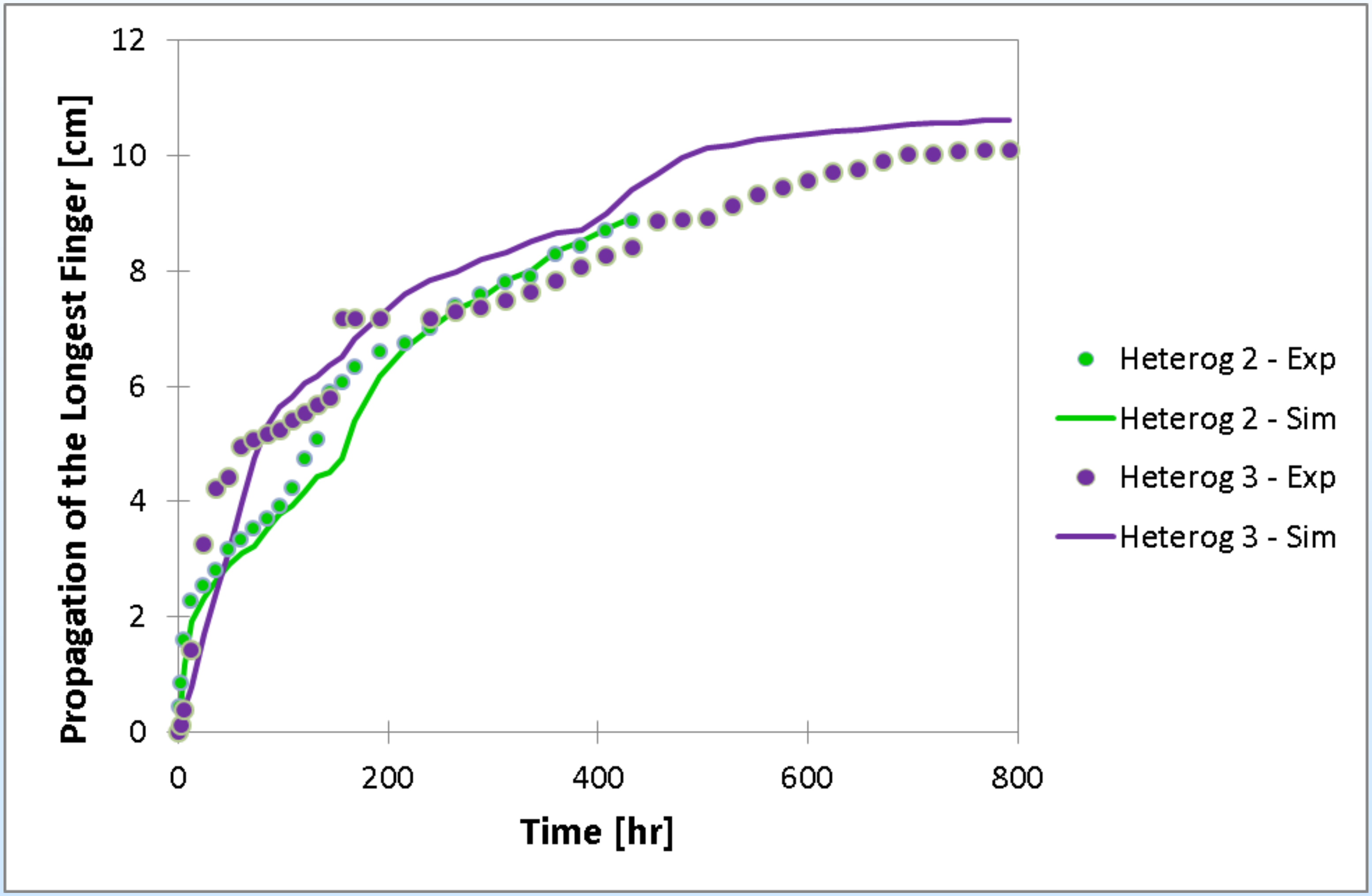


12-hour 3-day 9-day 18-day

Small Tank Experiments (*homogeneous*)

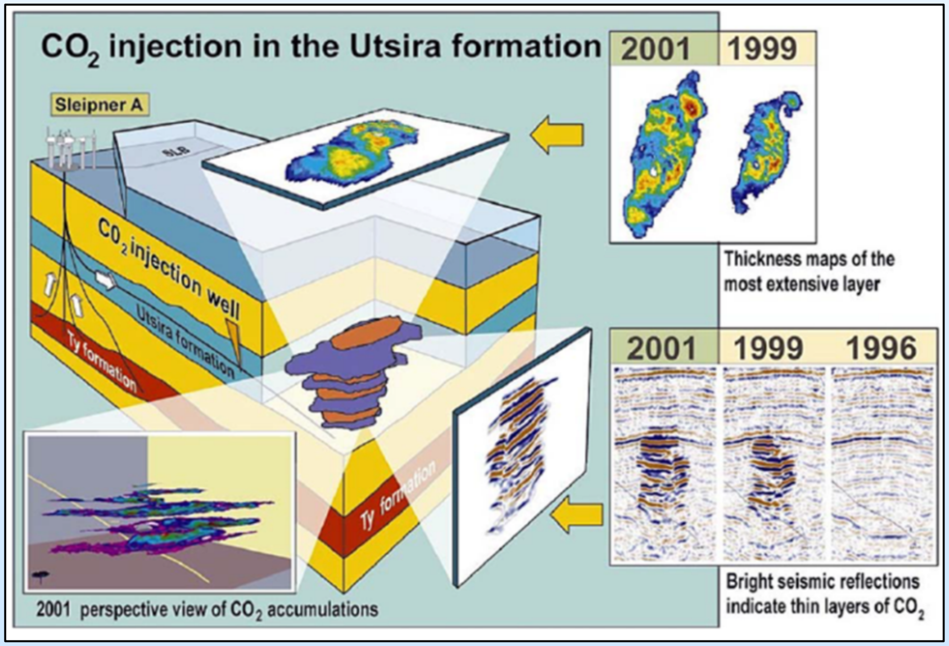


Small Tank Experiments (Heterogeneous)



Numerical Models for the Utsira Formation in the North Sea

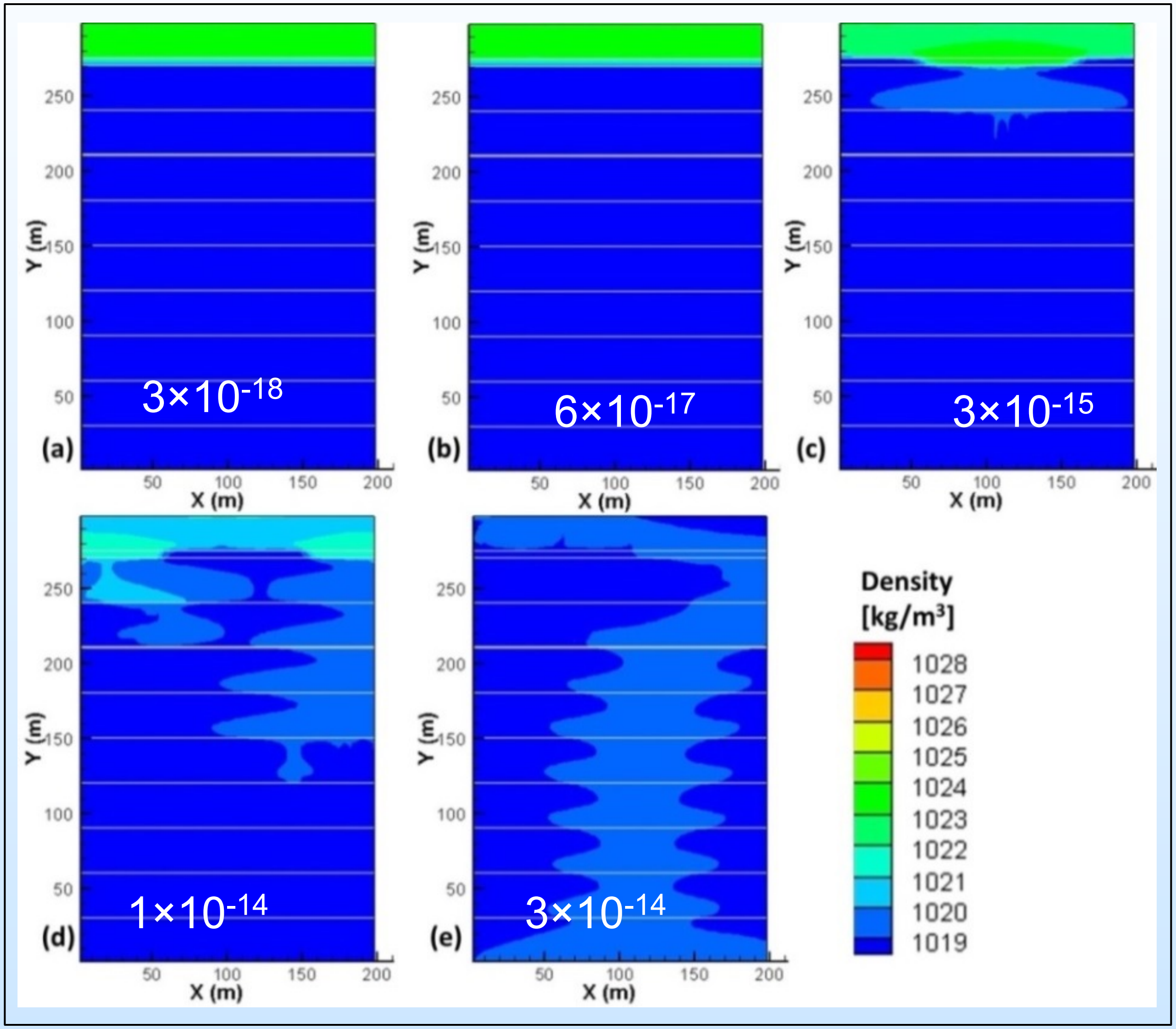
- Thickness, permeability and porosity of the Utsira sandstone are known.
- Uncertainty on shale layer geometries, distributions, and hydraulic properties [*Torp and Gale,* 2004].
- Simulations were carried out using the physical properties of $scCO_2$ and formation brine under sequestration conditions.



Seismic survey results showing the spreading of injected CO_2 in the Utsira formation [Torp and Gale, 2004].

Sensitivity of Shale Layer Permeability on Trapping

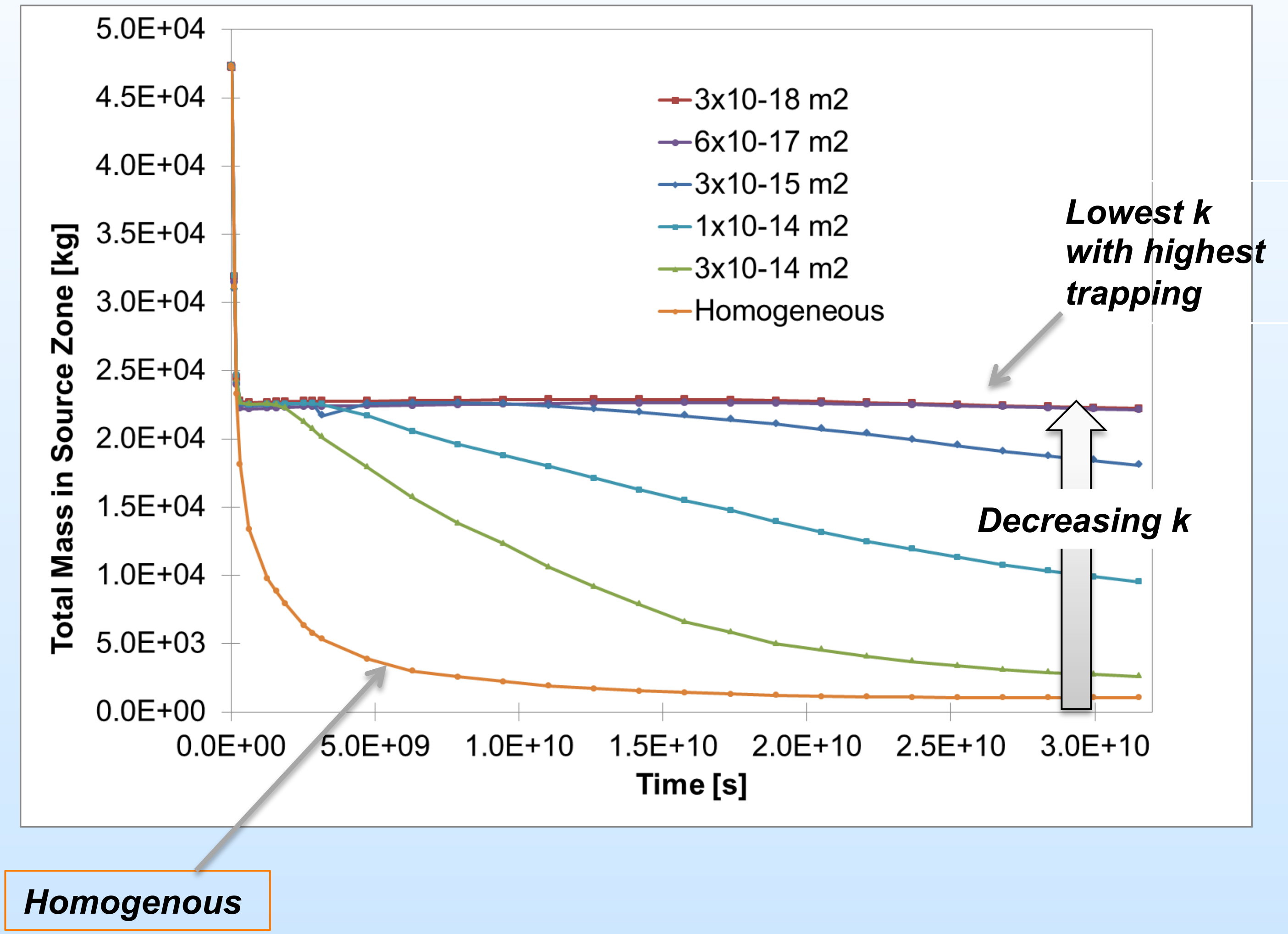
1000-year simulation results for different shale layer permeabilities [m^2]



- ✓ Lowest shale k retained more dissolved mass in the source zone

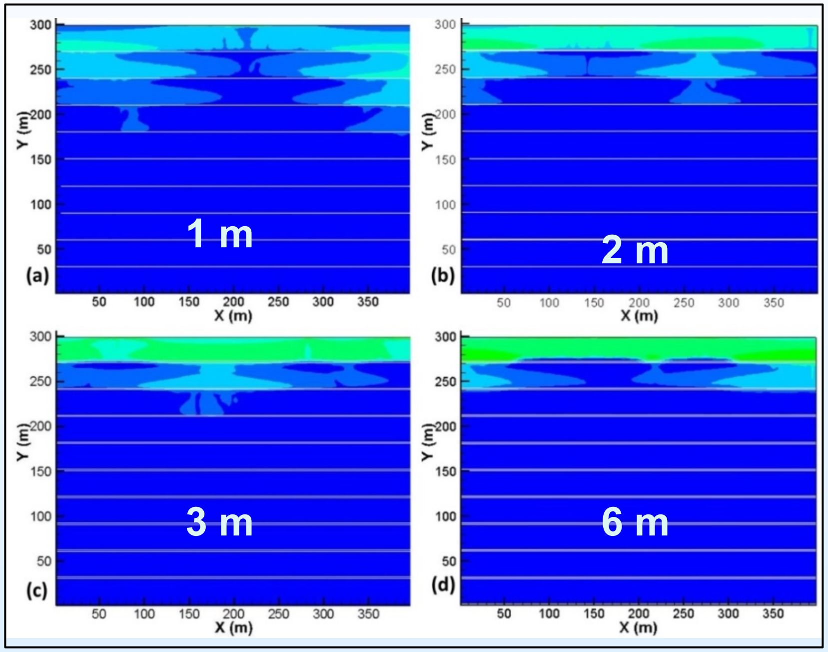
Sensitivity of Shale Layer Permeability on Trapping

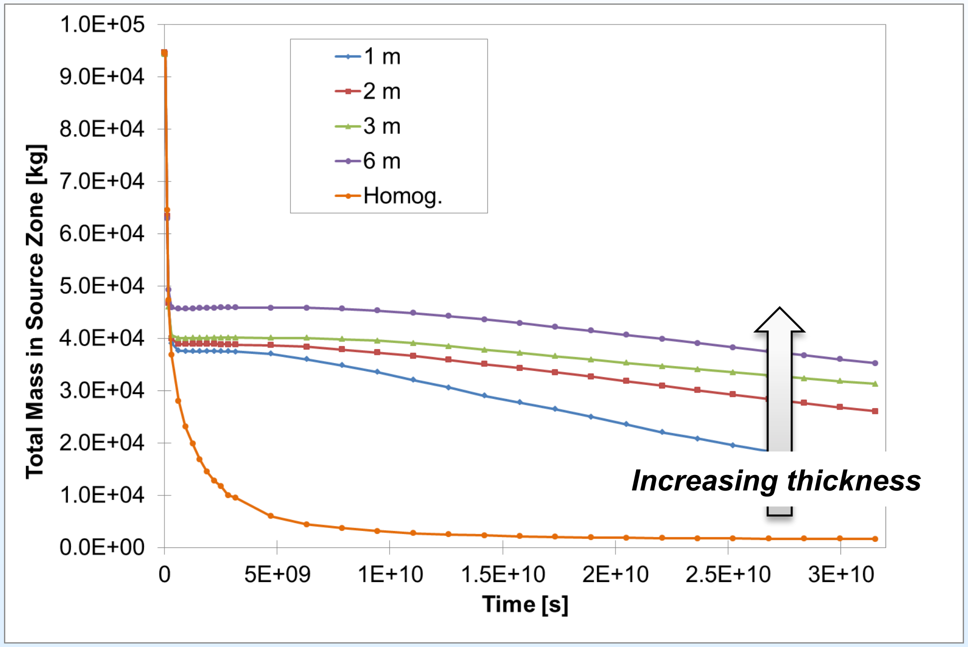
Variation of total mass remained in 12 m thick source zone with time for the different shale layer permeabilities.



Influence of Shale Layer Thickness on Trapping

1000-year simulation results for different shale layer thicknesses

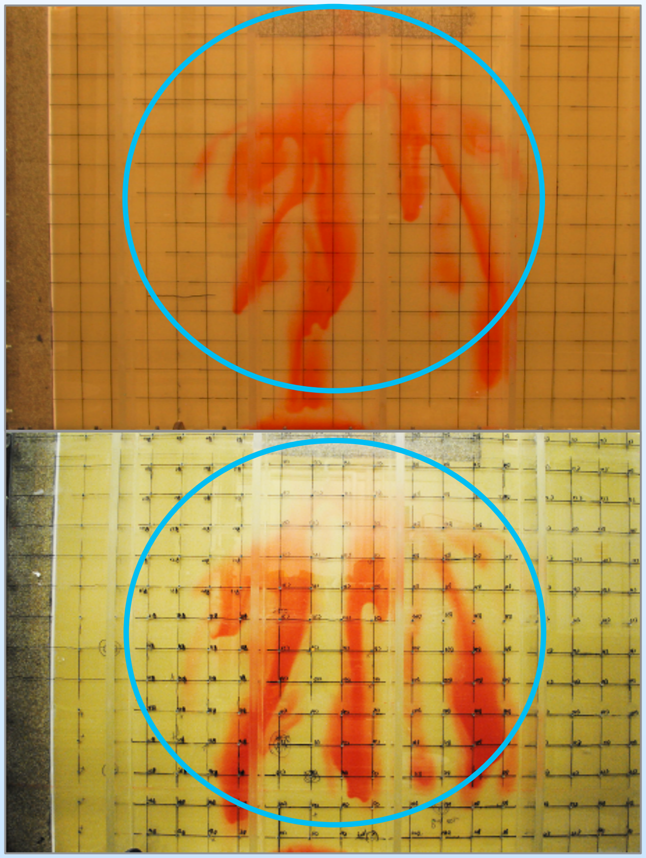




- ✓ Relatively higher permeability shale layers behaves like a homogeneous formation where convective mixing enhances the trapping in deeper parts for potential mineralization.
- ✓ Thick and lower permeability shale layers slow down the vertical spreading due to diffusive flow, which increases immobilized dissolved mass.
- ✓ Effective strategies can be developed to enhance trapping by taking the advantage of natural heterogeneity of the formation.

Homogeneous Large Tank Experiments

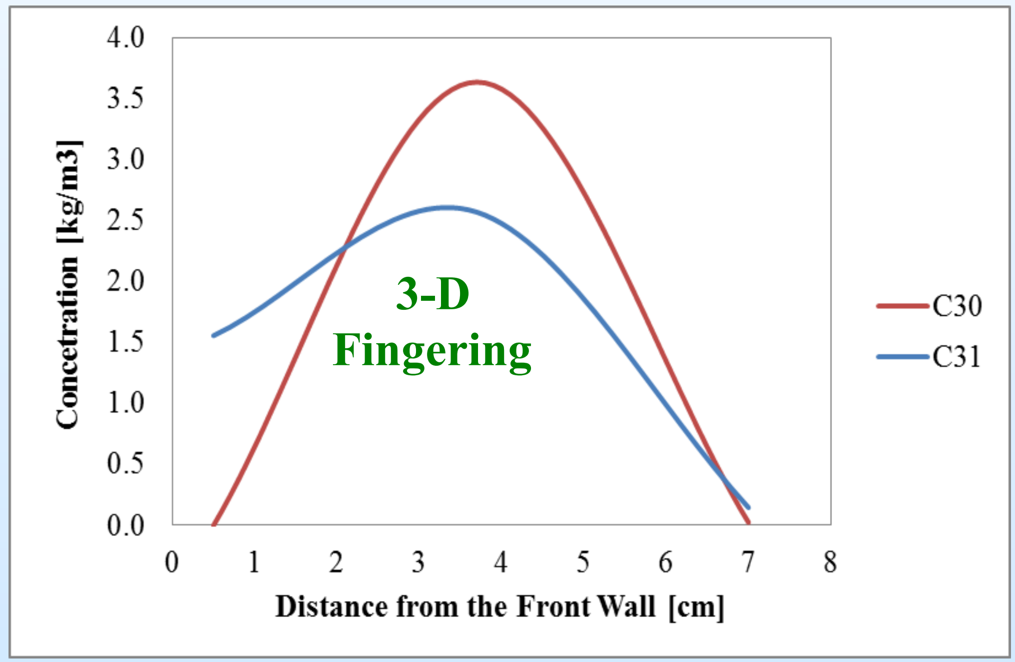
- ✓ Two homogeneous media large tank experiments were carried out using #40/50 sand. The source zone was packed with #8 sand. The injection was achieved using the syringe pump, and porous pipes were used to distribute the injected NaBr solution evenly in the source zone.



Front Wall

Back Wall

t= 60 hrs



Accomplishments (to date from project start)

- Task 2 – Experiments in intermediate-scale
  - ✓ Selected and tested surrogate fluids
  - ✓ Characterized laboratory sands to test existing constitutive models
  - ✓ Small tank experiments completed for testing capillary trapping and density-dependent fingers in homogeneous and simple heterogeneous systems
  - ✓ Performed large tank experiments for capillary and dissolution trapping in homogeneous systems
  - ✓ Setting up and packing large tank experiments with random heterogeneity
- Task 3 – Modeling
  - ✓ Simulated the two-phase flow in small tank and large tank experiments and compared the model results with experimental data
  - ✓ Developed a new multiphase flow solver (based on the Finite Volume method) for analysis of the experimental data and new constitutive models and non-equilibrium mass transfer
  - ✓ Developed a new code for analyzing heterogeneity: Computes connectivity based on invasion percolation algorithm. This code also involves algorithms to upscale two-phase flow parameters.
  - ✓ Developed a new hysteresis model and tested against few data sets

Findings (capillary trapping)

- The numerical models based on the classical two-phase flow theory were able to capture the main features observed during the migration of the $scCO_2$ surrogate fluid in the small tanks
- Single immiscible displacement experiments conducted in a quasi-two-dimensional flow cell allow for the measurement of a wide range of initial NWP saturations. This makes comparison of theoretical trapping models with actual trends observed in experimental data possible
- To minimize the occurrence of mobile NWP in the reservoir, a forced imbibition event such as a chase brine injection can eventually initiate a secondary drainage/imbibition cycle, leading to an enhanced NWP residual saturation at locations far-off from the injection well.
- Well-defined heterogeneity (existence of lower and higher permeability zones) enhances the capillary entrapment due to capillary barriers.
- Intermediate-scale laboratory experiments can be used to study plume behavior under different injection schemes

Findings (dissolution trapping)

- **Convection may not be the dominant mixing mechanism in layered systems with low permeability zones.**
- **Diffusive mixing increases the amount of dissolved mass immobilized in the low permeability zones.**
- **Thin and relatively higher permeability shale layers behaves like a homogeneous formation, where convective mixing is observed.**
- **Thick and lower permeability shale layers slow down the vertical spreading due to diffusion, which increases immobilized the dissolved mass.**
- **Lateral spreading observed at the boundary between high/medium and low permeability zones enhances the diffusion area and the diffused mass.**
- **Effective strategies can be developed to enhance trapping by taking the advantage of natural heterogeneity of the formation.**

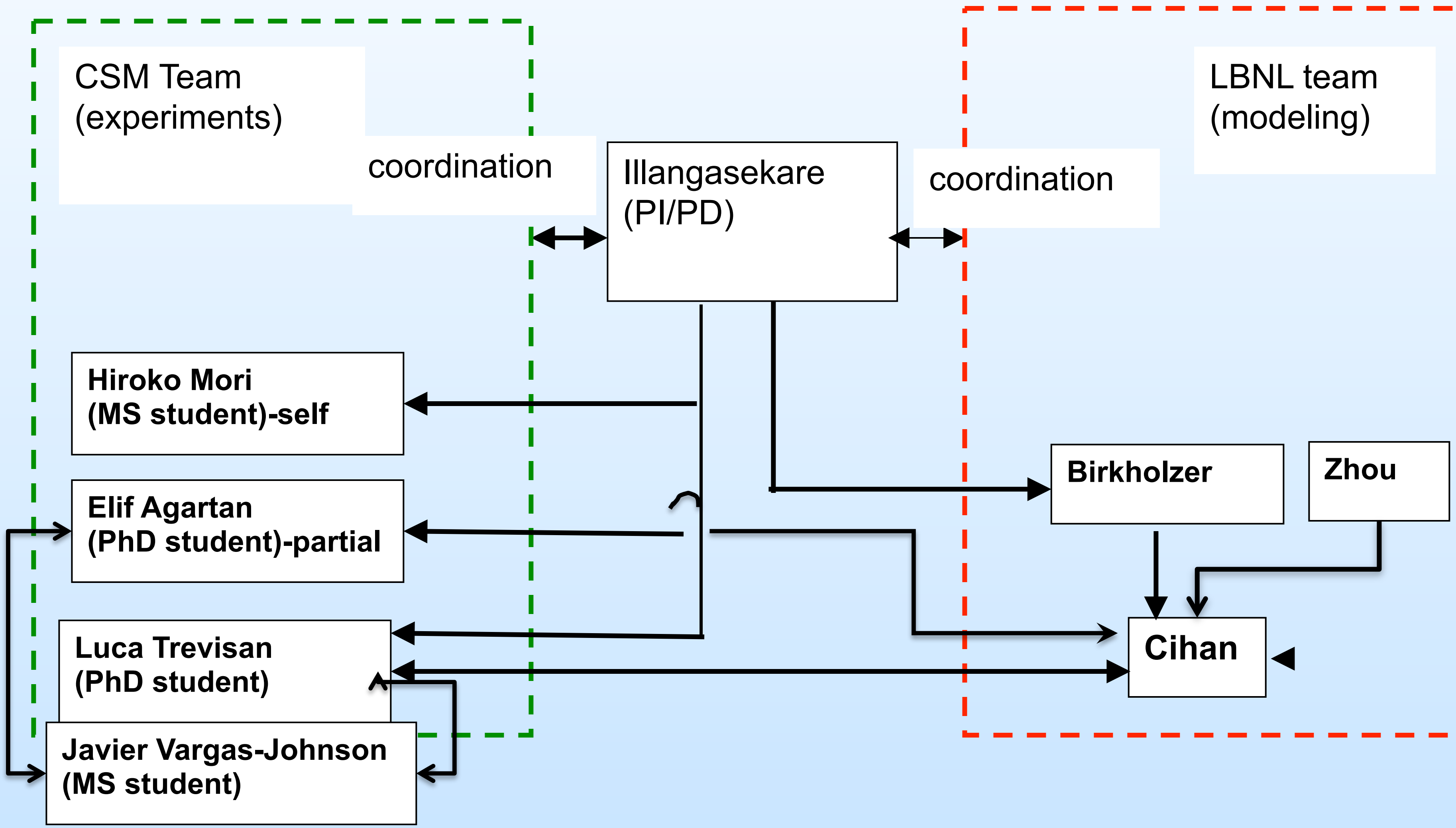
Future Efforts

- Complete large tank heterogeneous experiments on capillary trapping.
- Improve the numerical models by incorporating the validated constitutive models-issues of up-scaling
- Evaluate the ability of models to capture trapping during injection and forced imbibition.
- Use models to develop injection strategies to enhance trapping
- intermediate-scale heterogeneous experiment and models to examine the storage capacity of low permeability zones and the impacts of back diffusion
- Simulate dissolution of capillary trapped scCO2 for the determination of the effective mass transfer rate coefficient.

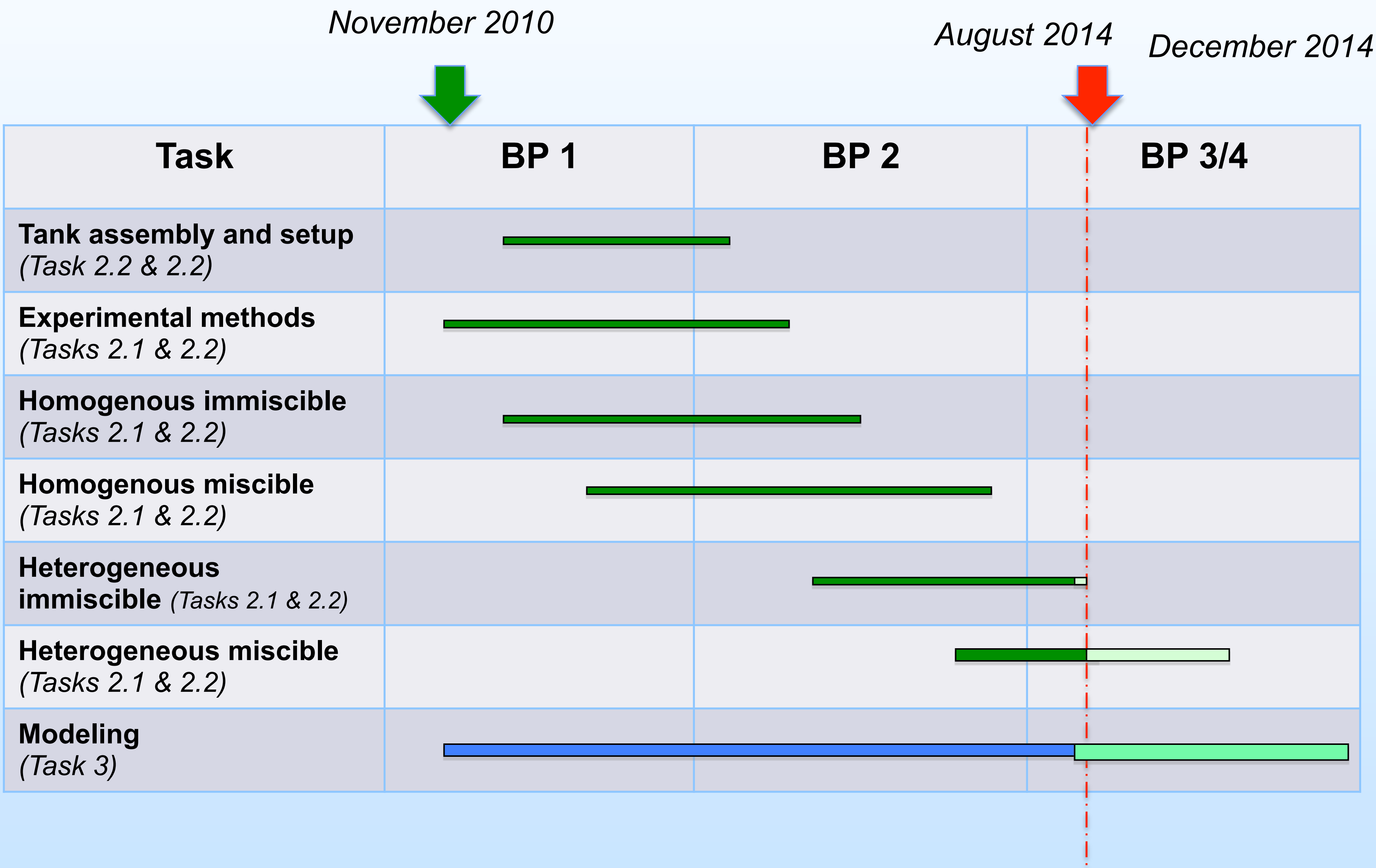
Appendix

– These slides will not be discussed during the presentation, but are mandatory

Organization Chart



Project Overview:
Schedule



Bibliography

Articles

- Trevisan, L., Cihan, A., Fagerlund, F., Agartan, E., Mori, H., Birkholzer, J.T., Zhou, Q. and Illangasekare, T. H., 2014. Investigation of mechanisms of supercritical CO_2 trapping in deep saline reservoirs using surrogate fluids at ambient laboratory conditions, *International Journal of Greenhouse Gas Control* (accepted)
- Trevisan, L., Pini, R., Cihan, A., Birkholzer, J.T., Zhou, Q. and Illangasekare, T. H. Experimental and numerical analysis of supercritical CO_2 trapping phenomena at the intermediate laboratory scale in well-defined heterogeneous porous media (in preparation)
- Agartan, E., L. Trevisan, A. Cihan, J. Birkholzer, Q. Zhou, and T.H. Illangasekare (2014a), Experimental Study on Effects of Geologic Heterogeneity in Enhancing Dissolution Trapping of Supercritical CO2, Water Resour. Res., (in review).
- Agartan, E., A. Cihan, J. Birkholzer, Q. Zhou, and T.H. Illangasekare (2014b), Effects of Lithology of Deep Layered Geologic Formations on Trapping of Dissolved CO2, Water Resour. Res., (to be submitted in August).
- Cihan, A., J. Birkholzer, T.H. Illangasekare and Q.Zhou, 2014. A Modeling Approach to Represent Hysteresis in Capillary Pressure-Saturation Relationship Based on Fluid Connectivity in Void Space, submitted in 2013. Water Resources Research, 50, 119-131.
- Mori, H.,T.Sakaki and and T. H. Illangasekare1Laboratory study of geological carbon sequestration using surrogate fluids: Measurement and scaling of capillary pressure and saturation relationships, , *International Journal of Greenhouse Gas Control* (accepted with revisions)

Bibliography

Conferences:

- Trevisan, L., A. Cihan, F. Fagerlund, E. Agartan, H. Mori, J.T. Birkholzer, Q. Zhou, T.H. Illangasekare. Investigation of multiphase modeling approaches for behavior of super critical CO2 in deep formations using analog fluids in the laboratory. Fall Meeting of the American Geophysical Union, San Francisco, CA, Dec 2013.
- Trevisan, L., Pini, R., Cihan, A., Birkholzer, J.T., Zhou, Q. and Illangasekare, T. H. Experimental and numerical analysis of supercritical CO_2 trapping phenomena at the intermediate laboratory scale in well-defined heterogeneous porous media. Gordon Research Conference, Lewiston, ME, Jul 2014.

- Agartan, E., Cihan, A., Birkholzer, J., Zhou, Q., and Illangasekare, T.H. (2014), Effects of Lithology of Deep Layered Geologic Formations on Trapping of Dissolved Supercritical CO_2, Gordon Research Conference and Seminar (GRC and GRS) on Flow & Transport in Permeable Media, 5-11 July 2014, Lewiston, ME, USA (Poster Presentation).
- Agartan, E., T.H. Illangasekare, A. Cihan, J. Birkholzer, Q. Zhou, L. Trevisan. (2013). A Fundamental Study of Convective Mixing of CO2 in Heterogeneous Geologic Media using Surrogate Fluids and Numerical Modeling. Proceeding from MODFLOW and More 2013: Translating Science into Practice, June 2-5, Colorado School of Mines, Golden, CO., USA, 467-471 (Poster Presentation).
- Cihan, A., Birkholzer, J., Bianchi, M., Zhou, Q., Illangasekare, T. H., Trevisan, L., “A New Connectivity-Based Upscaling Methodology for Multi-Scale Two-Phase Flow Processes in Heterogeneous Geological Formations”, the CCUS 12th Annual Conference, Pittsburgh, PA, May 13-16 2013.