SAND2010-7955P

Transport, Tracer Testing, and STAMMT-L

Sandia National Laboratories

KAERI Hydro Training

November, 2010

Sandia National Laboratories is a multi-program laboratory operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Outline

- Transport Basics
- Tracer Test Basics
- Multi-rate Transport (STAMMT-L)

Diffusion

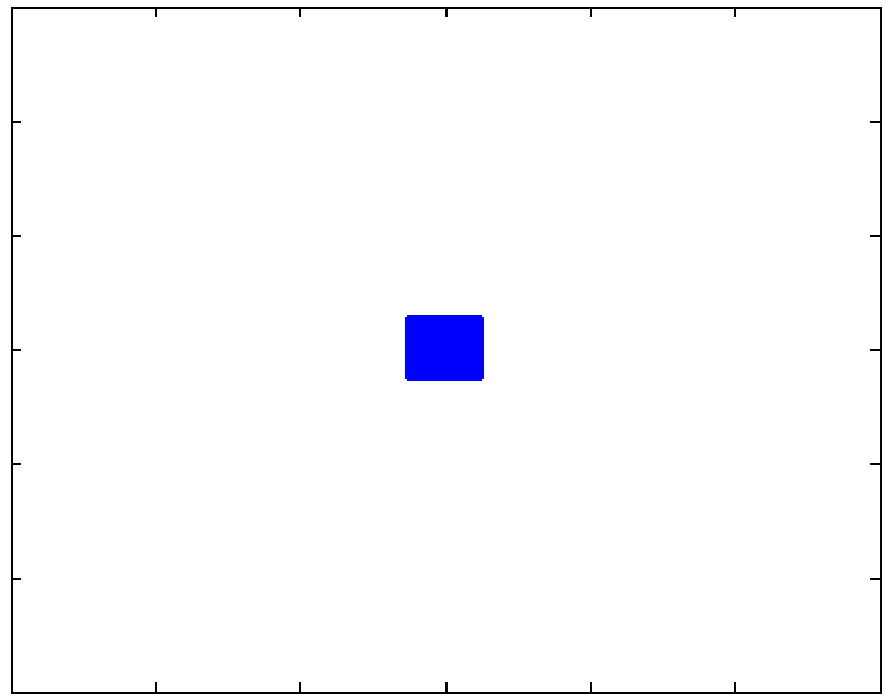
Spreading of a solute due to a concentration gradient

Fick's 1st law: Flux of solute is proportional to concentration gradient

$$F = -D\frac{\partial C}{\partial x}$$

D (L^2/T) is a function of the material(s) through which the solute is diffusing (e.g., open water is the aqueous diffusion coeff., D_{aq} *Around 8.0E-10* m^2/s)

F (M/T) is the rate of mass transfer per unit area



Analogy: Drop of dye in an aquarium

Diffusion: A Closer Look

$$F = -D\frac{\partial C}{\partial x}$$

Fick's first law

F	Mass flux of solute per area per time ($M/(L^2T)$)
D	Diffusion coefficient (L^2/T)
C	Solute concentration (M/L^3)
dC/dx	Concentration gradient $M/(L^3L)$

$$\frac{\partial C}{\partial t} = -\frac{\partial F}{\partial x}$$

Fick's Second Law: Mass conservation

Change in C with time = inflow - outflow

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2}$$

Combination of Fick's first and second laws: Diffusion differential equation

Diffusion: Details

Diffusion Coefficients:

Typically the aqueous or "free water" diffusion coefficient must be adjusted in some manner to account for the medium (e.g., unweathered granite vs. fault gouge)

$D_e = D_{aq}\tau$ Tortuosity of porous media ($\tau \sim 0.10$)

$D_e = D_{aq}\tau\phi$ Defined with porosity

Capacity of Medium:

Typical applications of diffusion models consider the medium into which the solute is diffusing to be "semi-infinite" – meaning there are negligble boundary effects from the opposite – closed end – of the material.

Definition of the length parameter – will consider in detail later

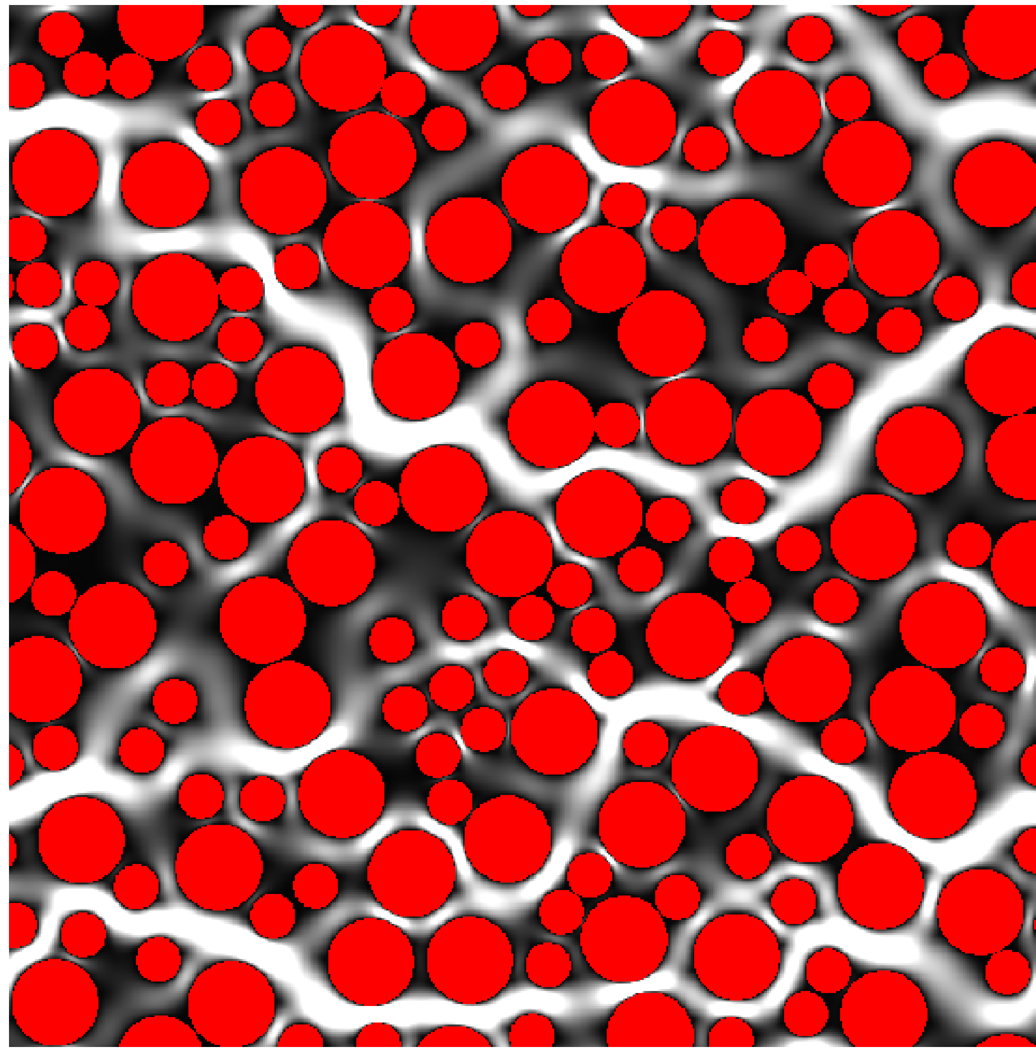
Mechanical Dispersion

Simulation of fluid velocities through porous media (35% porosity)

Relative fluid velocity

High

Low



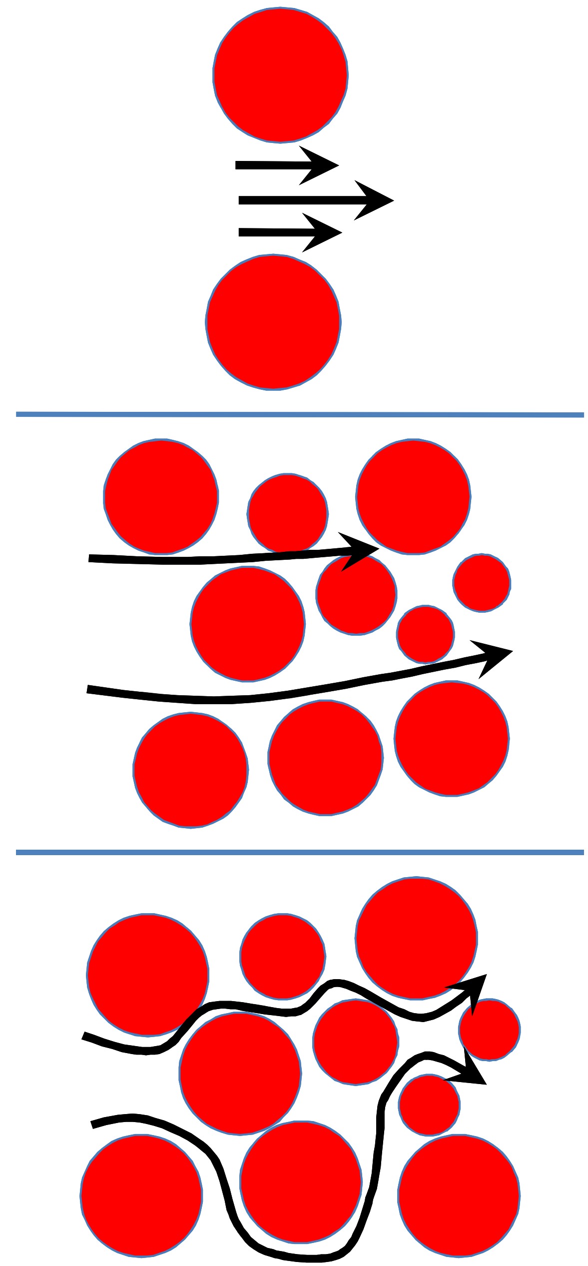
http://ciks.cbt.nist.gov/garbocz/paper32/fig14b.gif

Mechanical Dispersion: Longitudinal

Frictional forces in pore throats lead to *parabolic velocity distribution*

Variation in pore sizes leads to variation in pore velocity

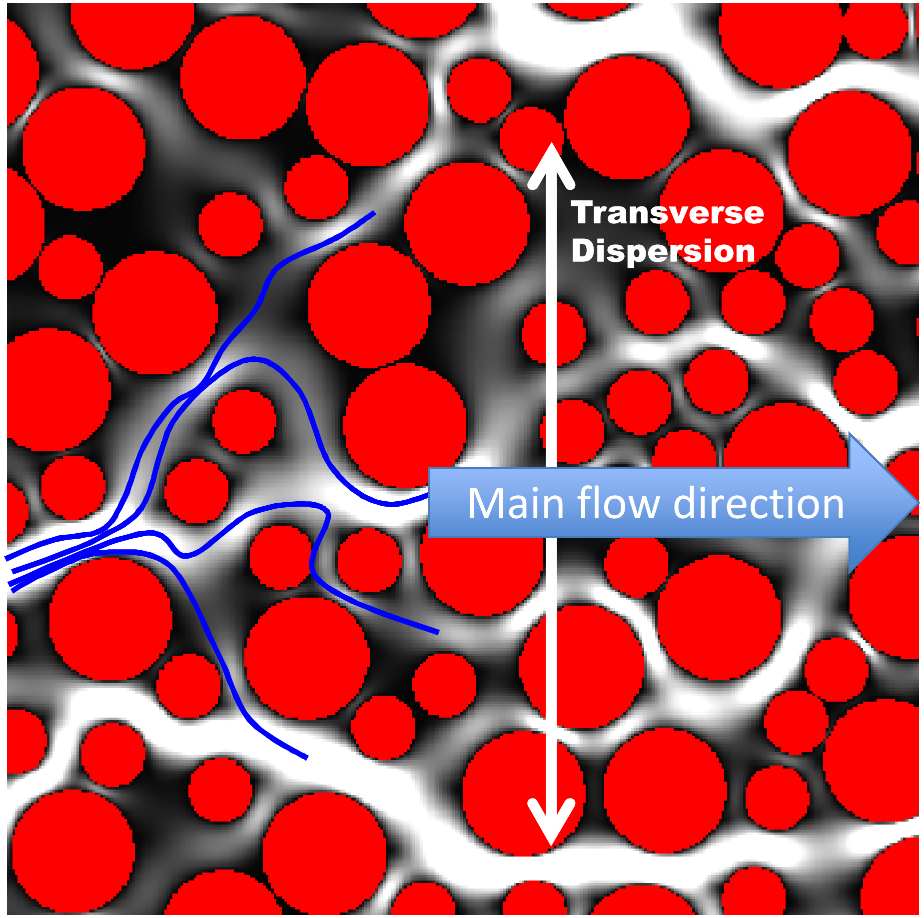
Variation in path lengths leads to variation in residence times



Mechanical Dispersion: Transverse

Variation in path lengths and velocities in the direction(s) orthogonal to the mean gradient leads to variation in flow path lengths and residence times

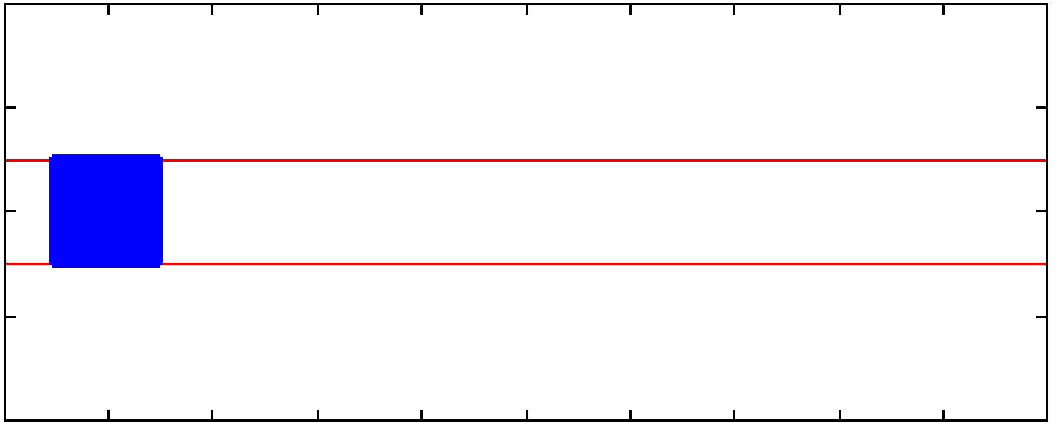
Summarize mechanical dispersion as a coefficient (dispersivity, units of length) acting on the mean velocity (1-D flow)



Dispersive Transport

Simple particle tracking model showing solute transport in a single fracture with matrix on top and bottom

Hydrodynamic dispersion is active, but <u>no mass-transfer</u> with matrix

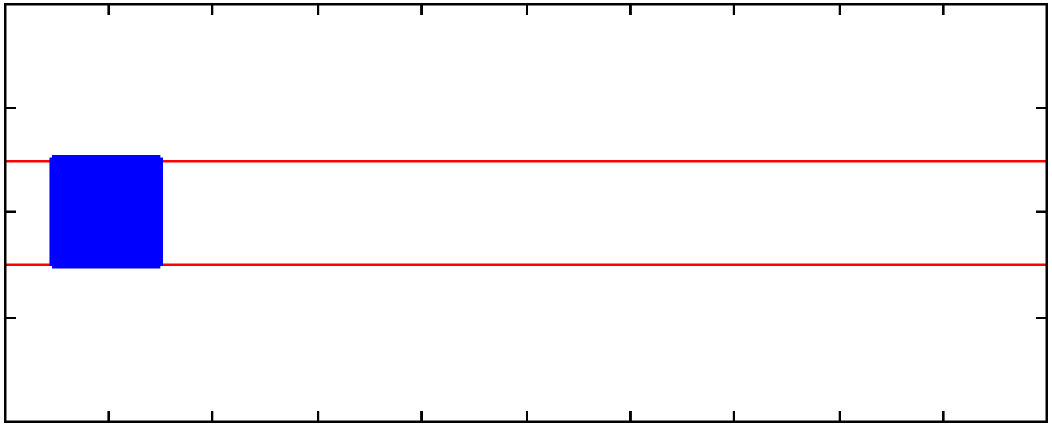


Also referred to as "single-porosity" transport

Dispersion and Diffusion

Simple particle tracking model showing solute transport in a single fracture with matrix on top and bottom

Hydrodynamic dispersion and mass-transfer are active



Also referred to as "dual-porosity" transport

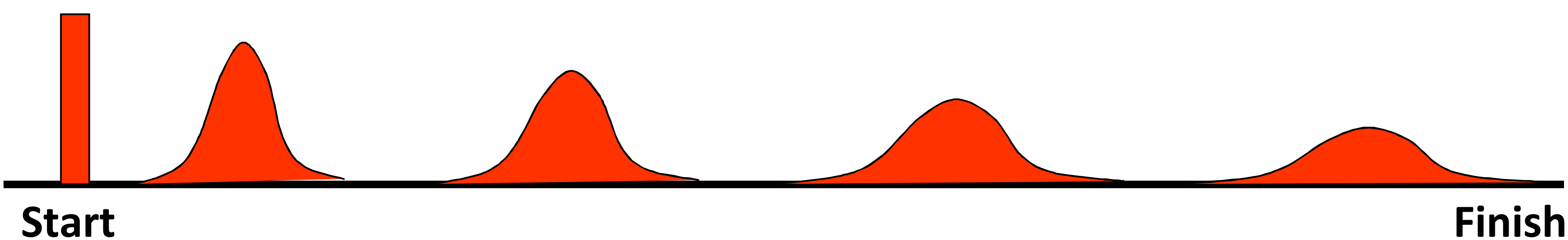
Hydrodynamic Dispersion

In most ground water flow problems, we lump the effects of mechanical dispersion and molecular diffusion into a single term: *hydrodynamic dispersion*

$$D_L = \alpha_L v_x + D^*$$

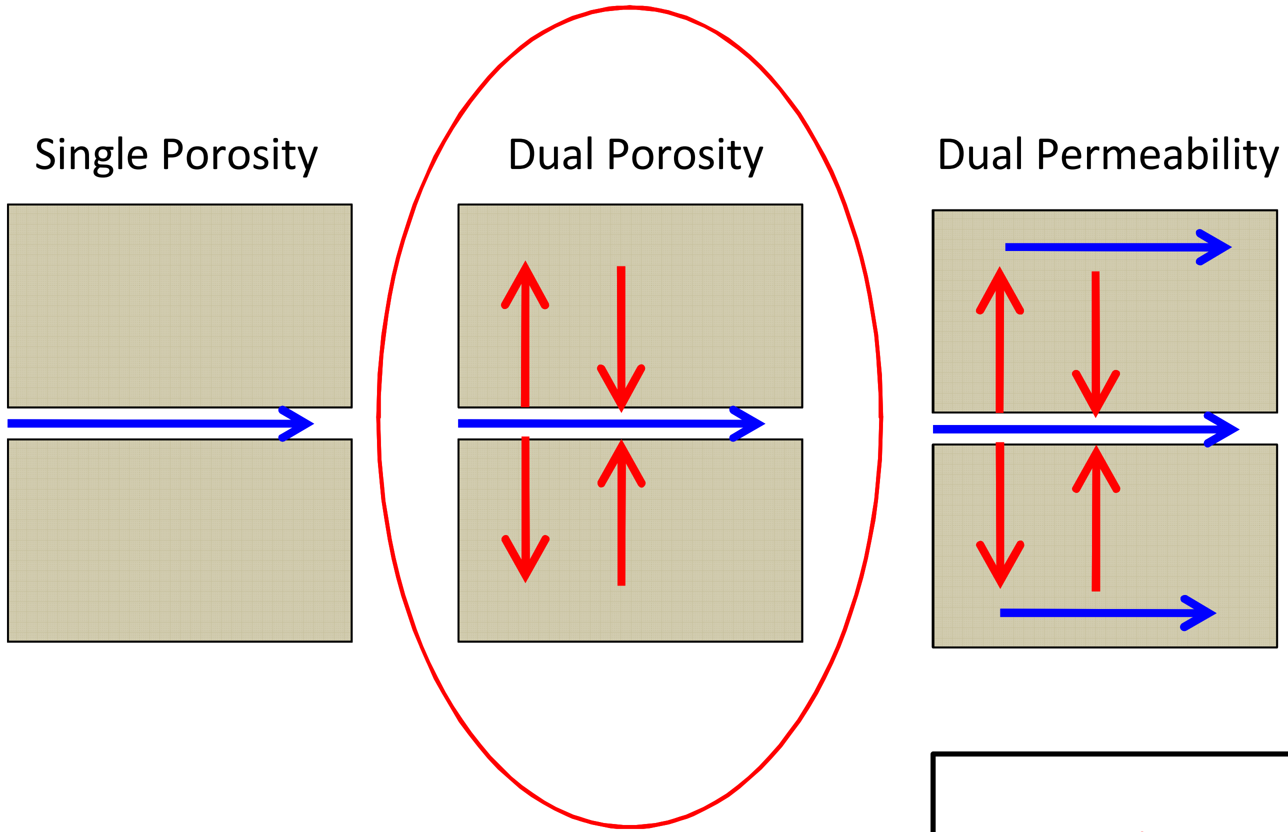
Units of L^2/T

Typically, $\alpha_L v_x >> D^*$



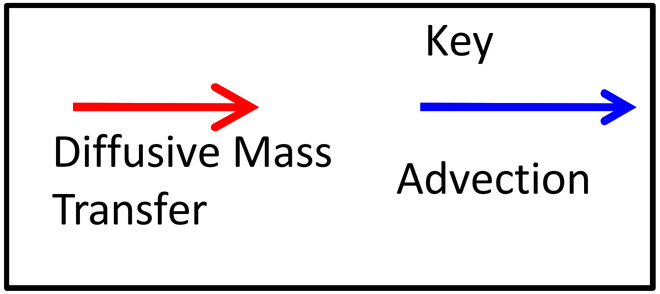
Analogy: a 10k running race

Conceptual Models of Transport



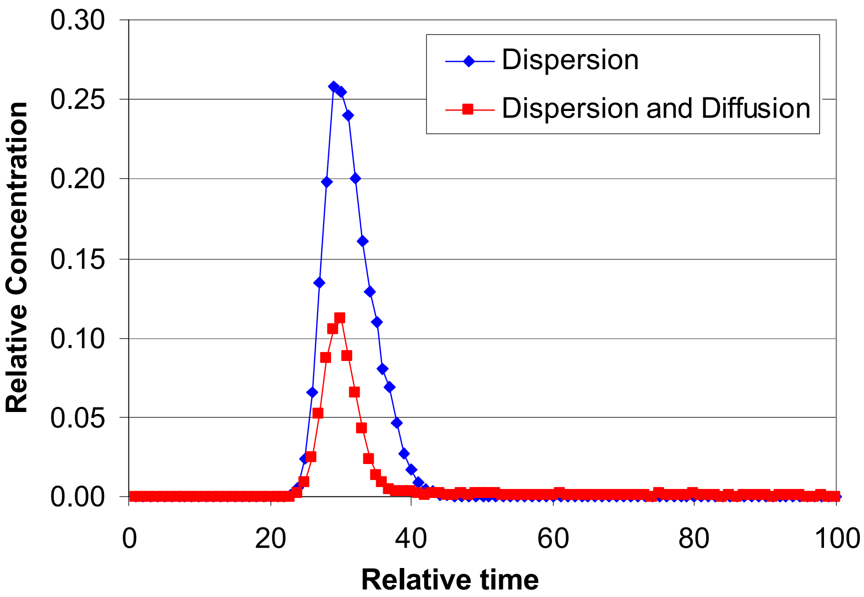
Mass transfer can be modeled as either:

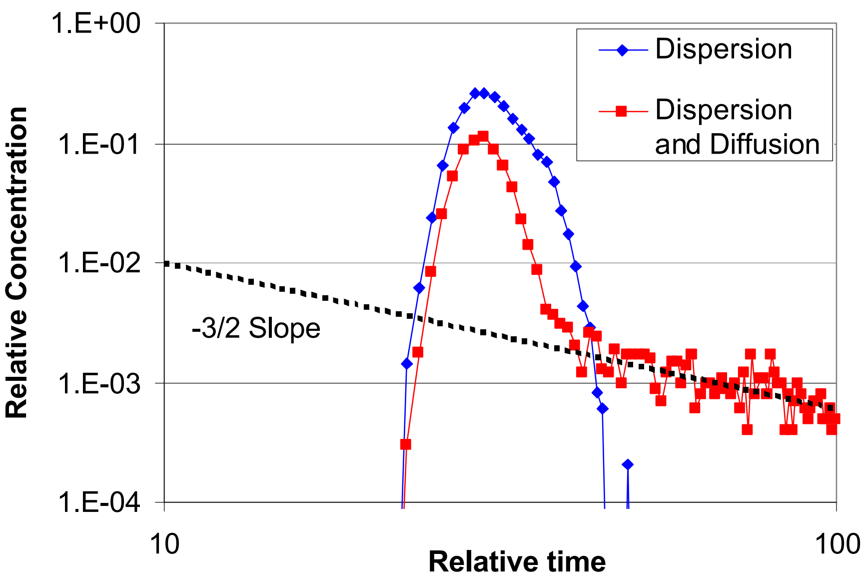
- Lumped parameter
- Spatially variable



Breakthrough Curves

The breakthrough curve is the plot of the concentration as a function of time at a downgradient location (e.g., pumping well).





- *To characterize tailing behavior, you* ***must examine results in log-log space***
- *-3/2 slope is characteristic of diffusion into an infinite medium*

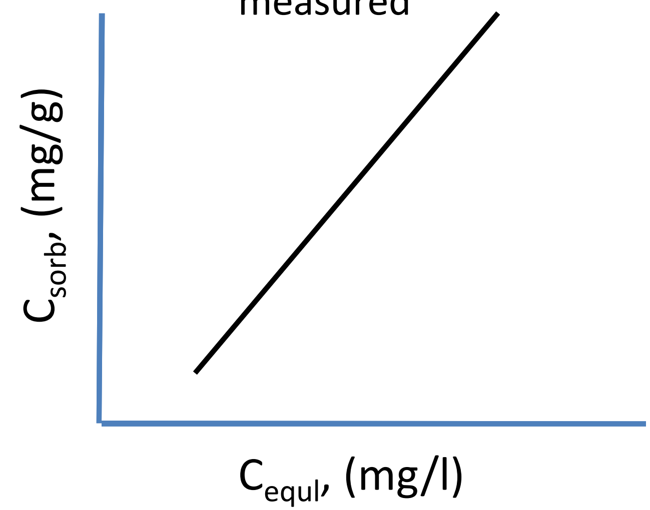
Sorption

Discussion so far has covered "conservative" solutes. Solutes that adsorb to a material are considered "reactive"

Mechanistic process described by coefficients of fit to experimental data

$$K_d = \frac{\partial C_{sorb}}{\partial C_{equil}}$$

Batches of solute and material are put together and then solute on the material and solute in the liquid are measured



The distribution coefficient, K_d (L^3/M), is the ratio of the amount of solute sorbed per mass of solid material to the equilibrium concentration of solute

Here only looking at case of linear relationship, the *Freundlich isotherm*

Bethke and Brady, 2000, How the Kd approach undermines ground water cleanup, Ground Water

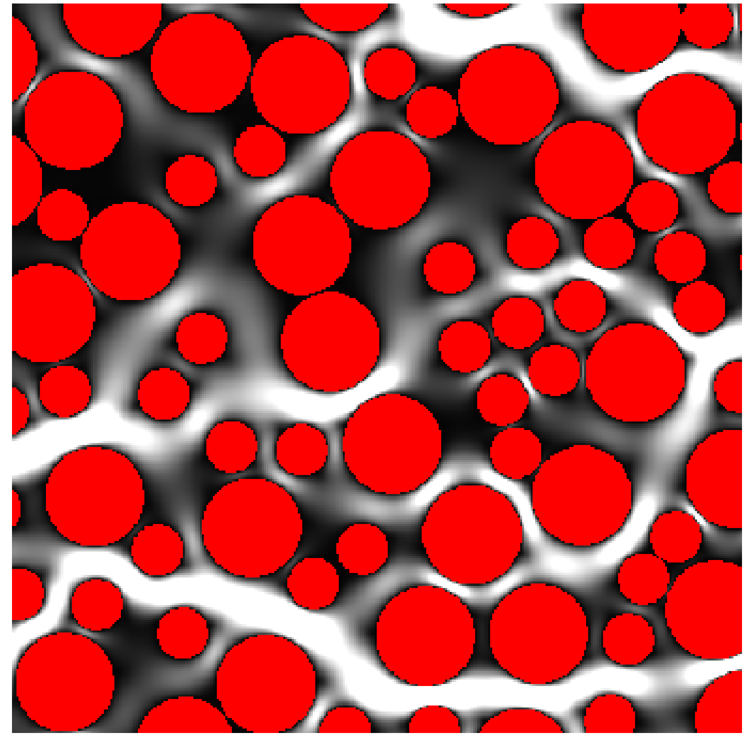
Retardation

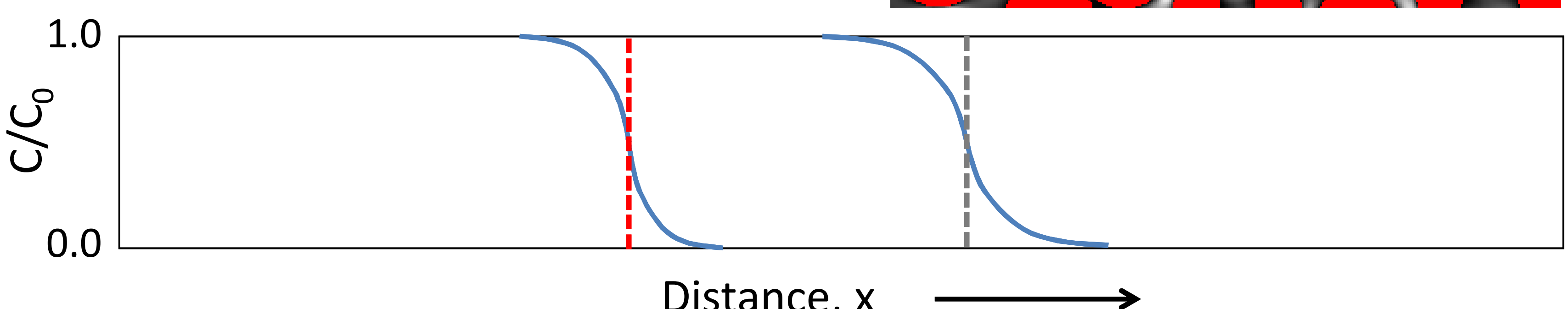
K_d is used to calculate the retardation coefficient.

$$R = 1 + (\rho_b / \phi) K_d$$

R = retardation coeff (-)
ρ_b = bulk density of matrix material (M/L^3)
ϕ= porosity (-)

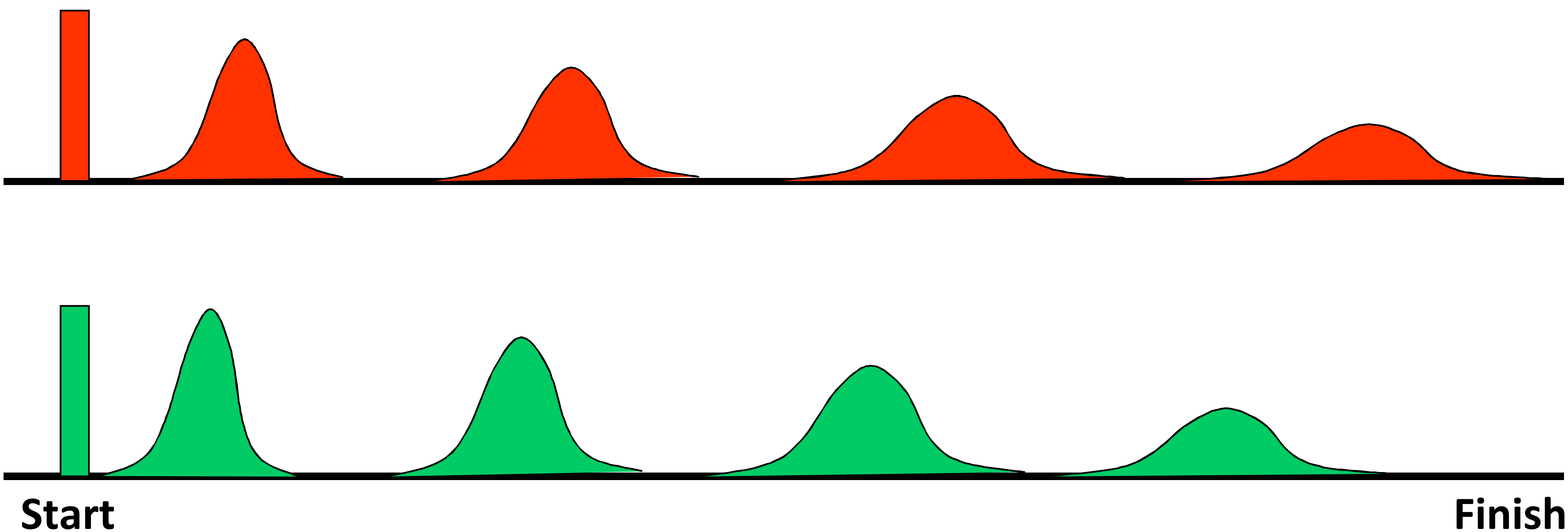
$V_{solute} = v_x/R$





Sorption Analogy

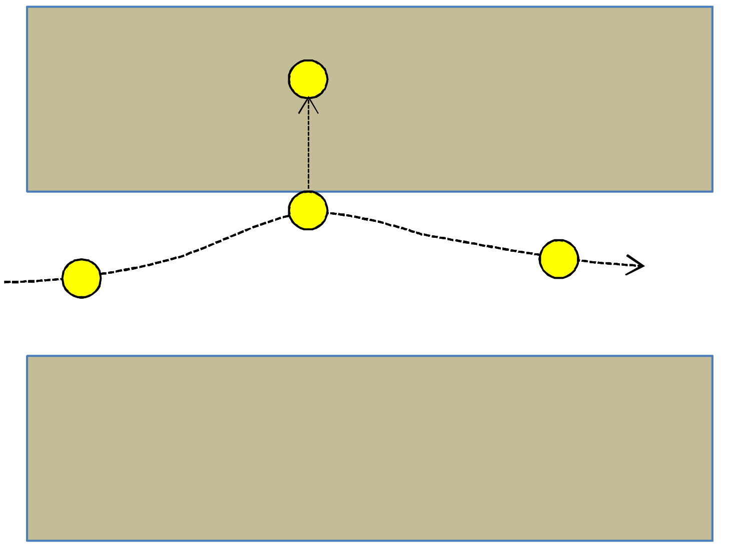
Adherence of solute onto a surface (fracture or pore)



Analogy: A 10k running race with faulty shoe laces

Sorption in Fractured Materials

Define a fracture surface area distribution coefficient, K_A



$$R_f = 1 + K_A A$$

K_A = mass of solute per unit area of fracture wall per unit concentration of solute in solution $(M/L^2)/(M/L^3) = L$
A = fracture area per volume of fracture (1/L)

Sorption in fractured media becomes complicated as the surface sites can be accessed from the advective domain, but the majority of sites in the matrix are accessed through diffusion

Transport Background Summary

Three main processes covered here:

Dispersion	Spreading of solute front due to pore-scale heterogeneity
Diffusion	Brownian motion at the molecular scale
Sorption	Sticking of solute under equilibrium conditions

All three processes have been adapted for transport in fractured rock

Dispersion	Variations in fracture aperture create variations in velocity
Diffusion	Provides mass-transfer between advective & diffusive domains
Sorption	Retardation factors for both fracture surface and matrix

Analytical solutions for some scenarios used in hands-on exercises

Discussion so far has focused on transport behavior in single fracture

Tracer Testing

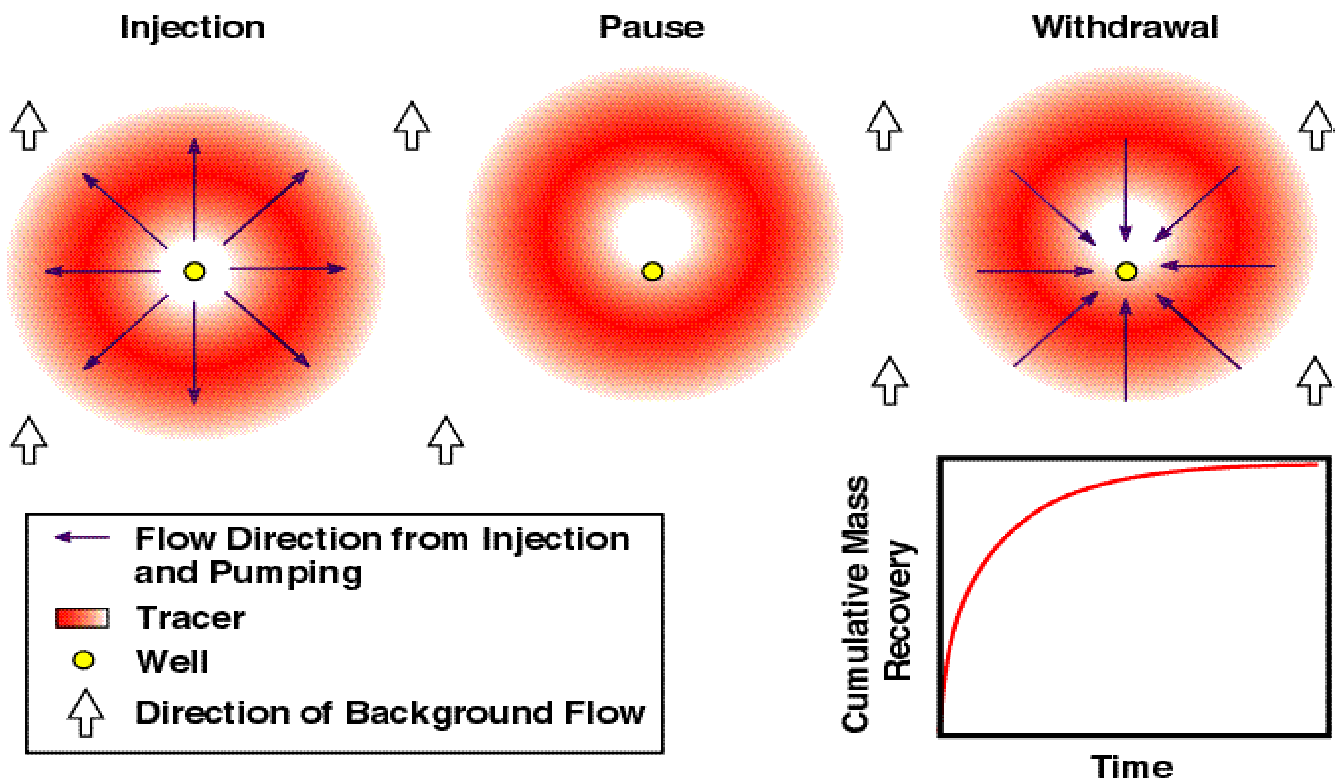
Overview

- Types of Tracer Tests
  - Single-well injection-withdrawal (push-pull)
  - Dipole
    - Balanced
    - Unbalanced
  - Convergent flow
  - Natural gradient
- Tracer Test Examples
- Use of Tracers in PA

Single-Well
Injection-Withdrawal Tracer Tests

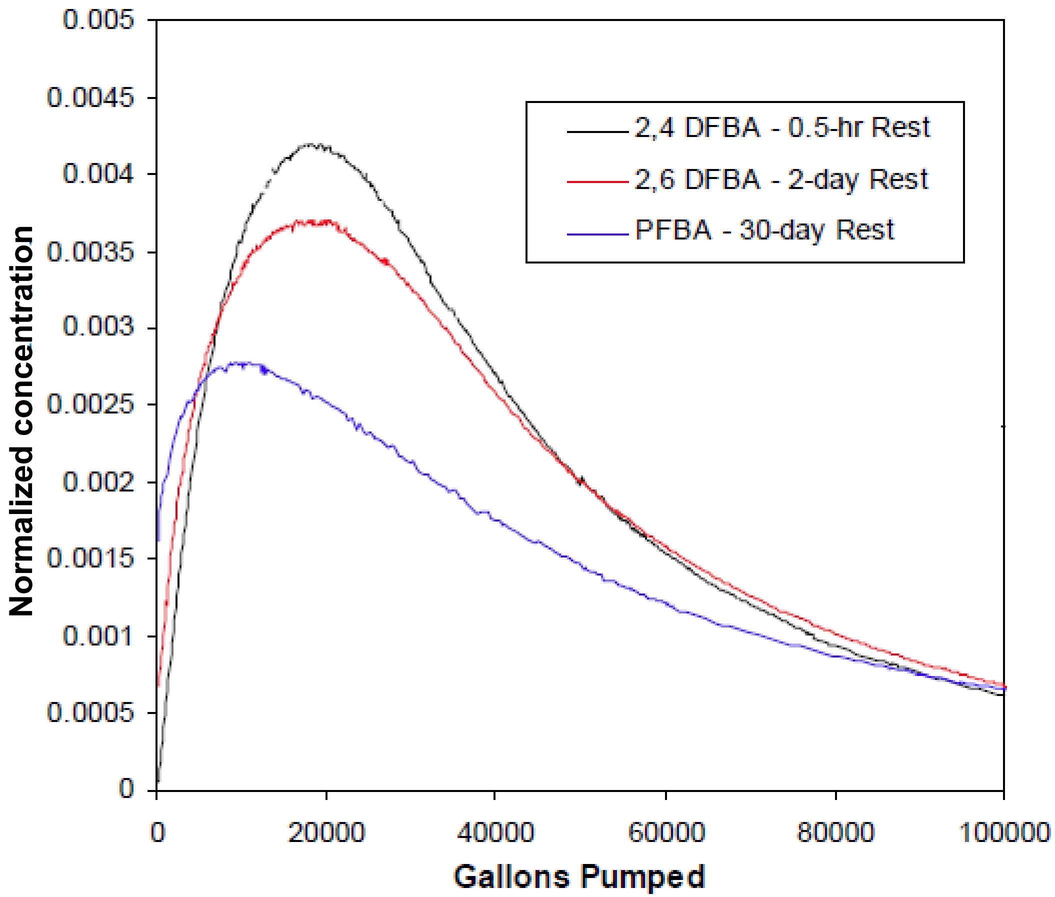
Three steps:

1. **Inject tracer followed by chaser**
2. **Pause to allow tracer to drift**
3. **Pump to recover tracer**



Single-Well Injection-Withdrawal Tracer Tests

Effect of pause period on tracer-recovery curve



Longer pause leads to:

- Faster initial recovery because drift brings tracer back to well
- Lower peak concentration because more dispersion and diffusion have occurred

Figure from Reimus and Robinson (2004)

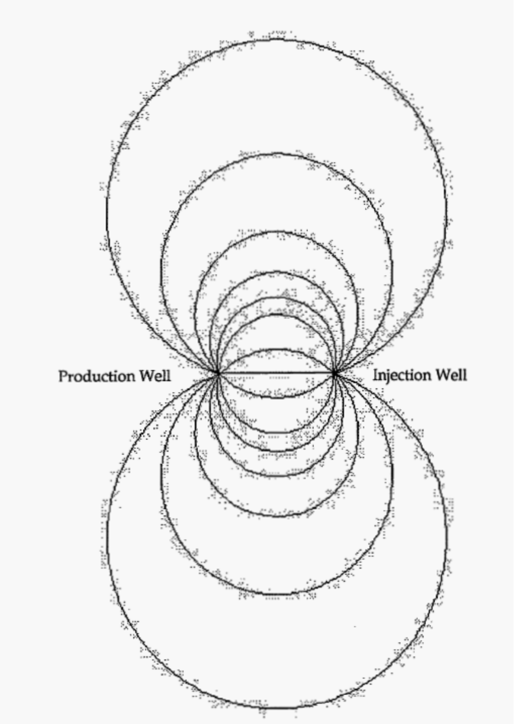
Strengths and Weaknesses of Single-Well Injection-Withdrawal Tests

- Strengths:
  - Best test for demonstrating multirate matrix diffusion
  - Low requirements (wells, equipment, tracers, analyses, time, money)
- Weaknesses:
  - Provides little information on heterogeneity and dispersion
  - Insensitive to advective (transport) porosity, due to uncertain test volume size.

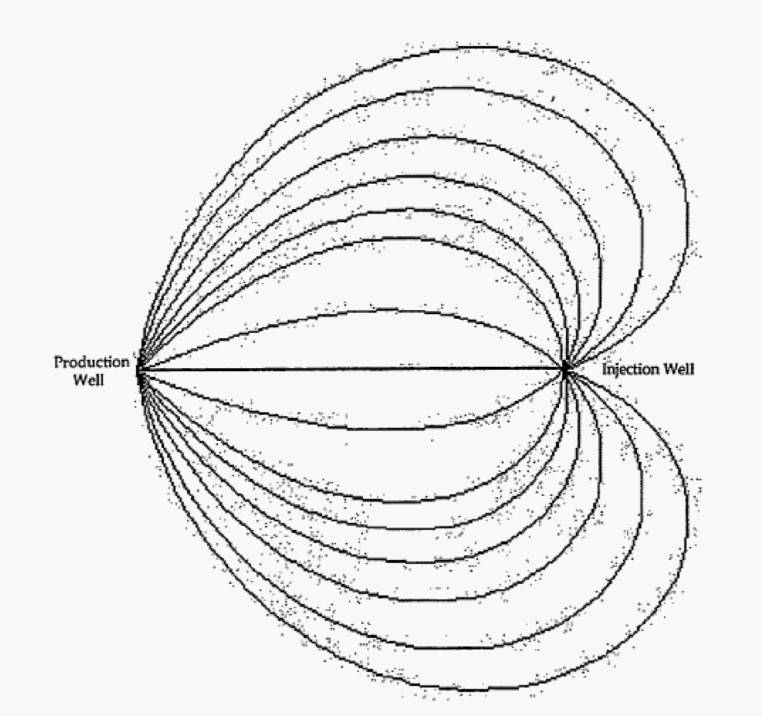
Dipole Tracer Tests

- Create stable flow field by continuously pumping from production well while injecting into injection well
- Inject slug of tracer in injection well and monitor tracer concentration at production well and possibly intermediate wells

Balanced



Unbalanced

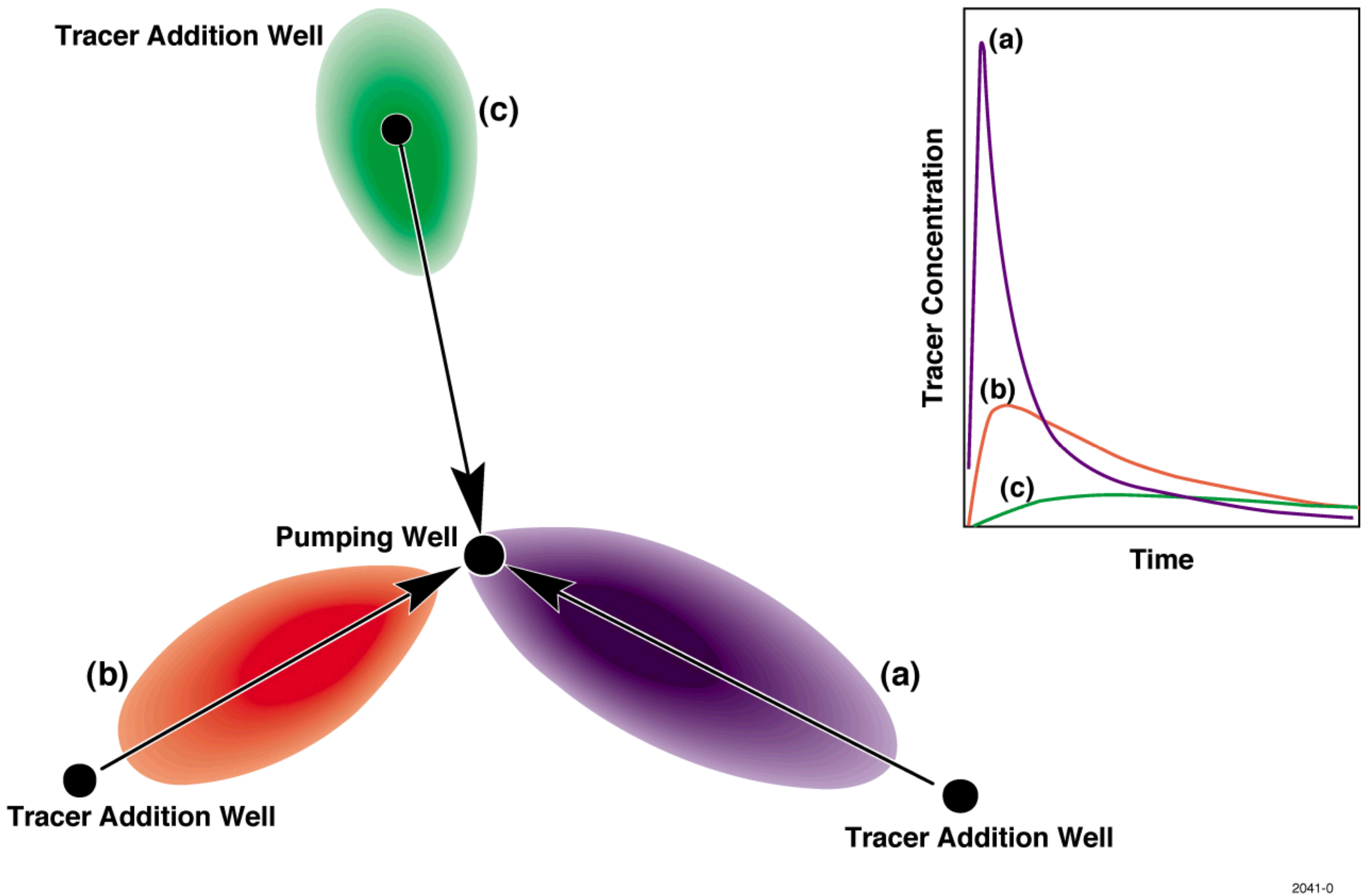


Figures from Reimus (1996)

Strengths and Weaknesses of Dipole Tests

- Strengths:
  - Best test for defining advective porosity
  - No water disposal issues
  - Tests larger volume of aquifer than two-well convergent-flow test (especially balanced dipole)
- Weaknesses:
  - Relatively insensitive to multiple rates of diffusion, due to typical high rates of flow between wells
  - Only tests pathways connecting the two wells
  - Provides no information on directional properties/variations

Convergent-Flow Tracer Tests



Strengths and Weaknesses of Convergent-Flow Tests

- Strengths:
  - Best test for defining advective porosity
  - Provides most information on three-dimensional variation in transport properties (heterogeneity)
- Weaknesses:
  - High requirements (wells, equipment, tracers, analyses, time, money)
  - Relatively insensitive to multiple rates of diffusion
  - Small volume of aquifer sampled from each tracer-injection well
  - Large volumes of water to be disposed

Natural Gradient Tracer Tests

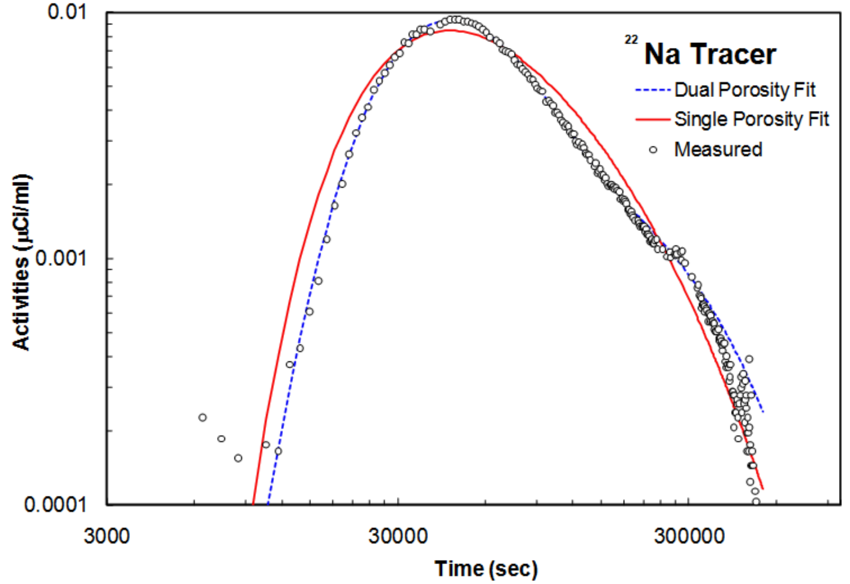
- Establish tracer source
  - One or more injection wells
  - Known contaminant source
- Construct extensive well field down gradient of source to map out plume distribution with time
- Typically performed in unconfined (water table) aquifers
- Typically involve multilevel sampling to map plume in three dimensions
- Sampling performed with minimal disturbance to flow field

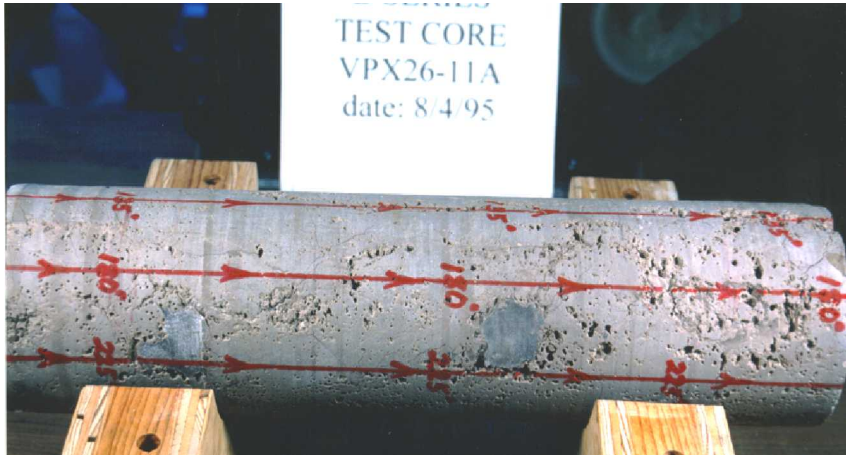
Strengths and Weaknesses of Natural Gradient Tracer Tests

- Strengths:
  - Best test for determining how contaminants might actually move
  - Provides most information on ambient velocities, longitudinal and transverse dispersivities
- Weaknesses:
  - Long time to complete
  - MANY monitoring wells required to delineate plume
  - Requires extensive sampling

Column Studies

- Controlled conditions
- Smaller scale
  - Benefits:
    - shorter test durations
    - less effluent
  - Shortcoming:
    - doesn't sample large-scale heterogeneity
- Can use tracers that wouldn't be feasible in field settings (e.g., hazardous, radioactive)
- Can destructively and comprehensively analyze sample post-testing

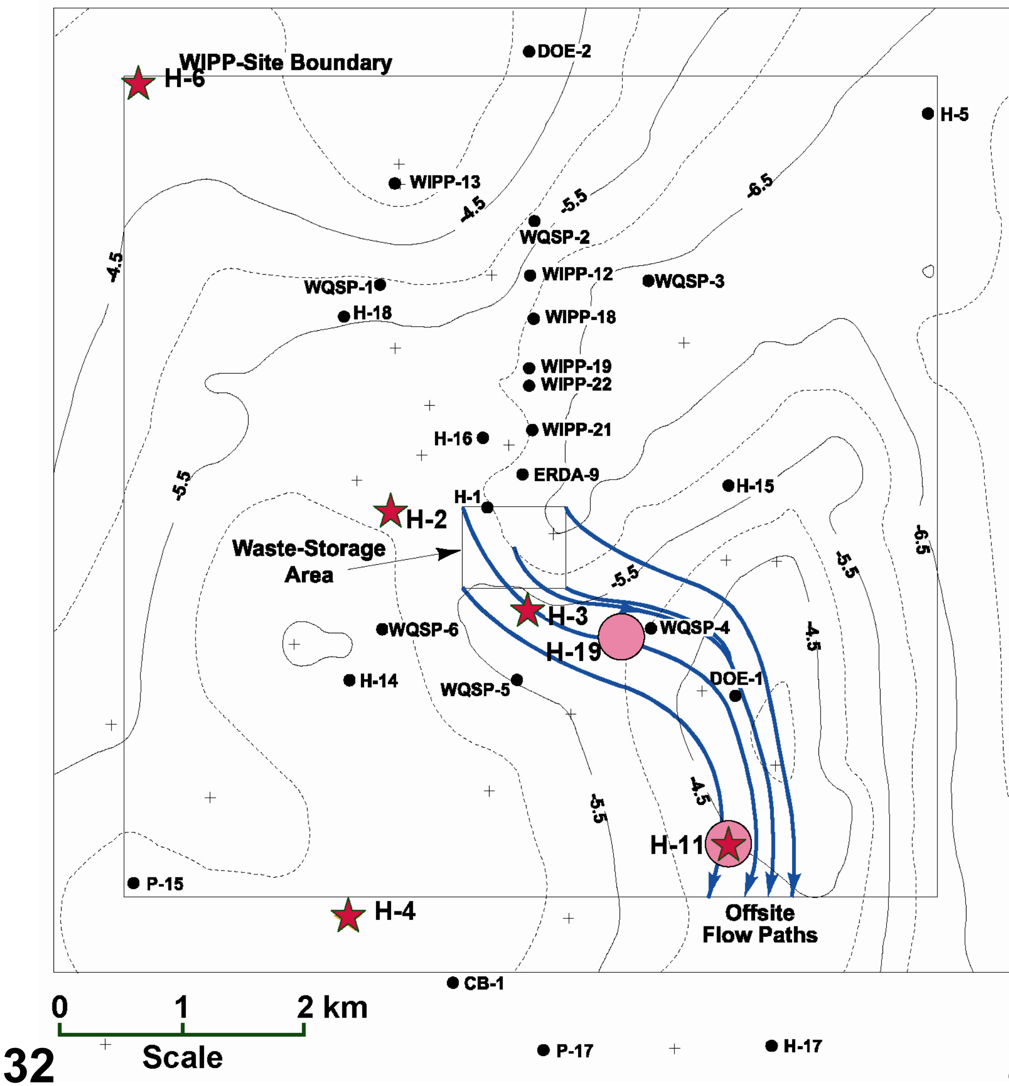




Lucero, D.A., G.O. Brown, and C.E. Heath, 1998. *Laboratory Column Experiments for Radionuclide Adsorption Studies of the Cuelbra Dolomite Member of the Rustler Formation*, SAND97-1763

Example Tracer Tests

Tracer Tests at the WIPP Site



- 1980 – 1986 Tracer Tests
  - 5 Locations
  - Two Types of Tests
    - Convergent-flow tests
    - Two-well recirculating (dipole) tests
- 1995-1996 Tracer Tests
  - 2 Locations
  - Two Types of Tests
    - Convergent-flow tests
    - Single-well injection- withdrawal tests

Location of 1980-1986 Tracer Tests

Location of 1995-1996 Tracer Tests

Observation Well

+ Pilot-Point Location

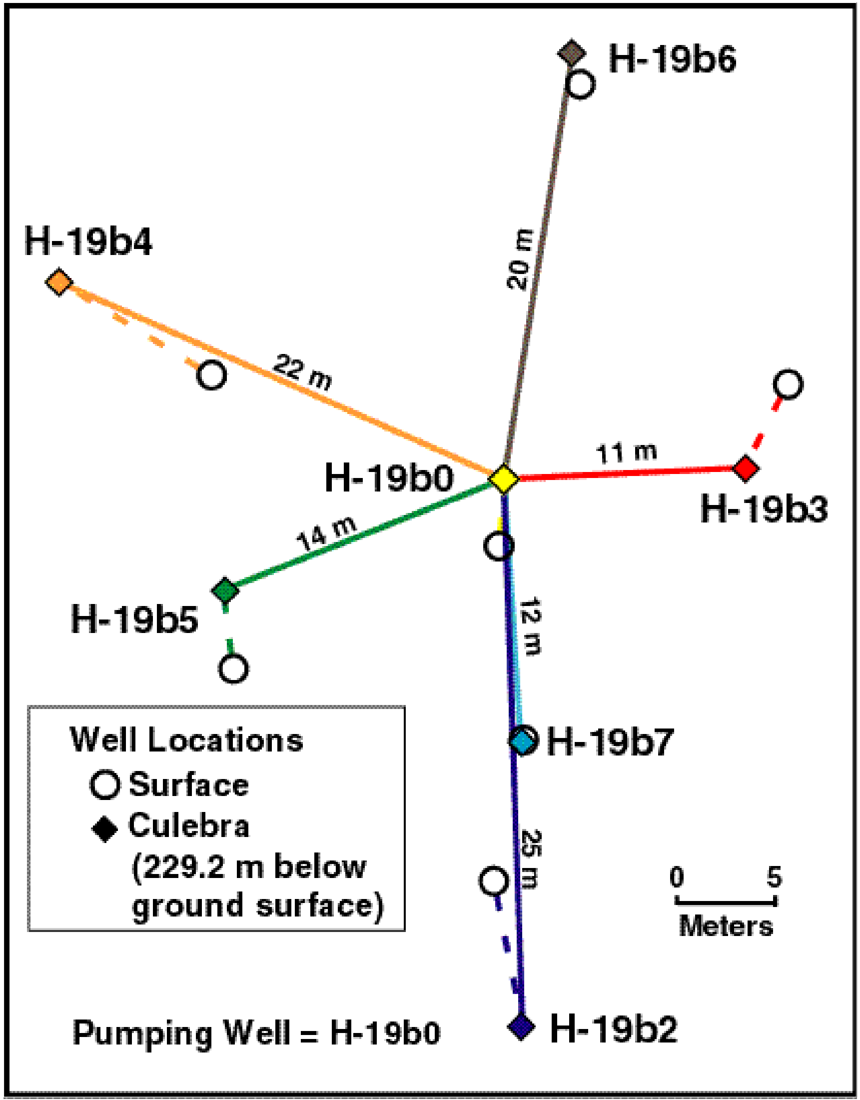
Transmissivities in $\log_{10}$ m^2/s
Contour Interval 0.5 $\log_{10}$ m_2/s

Meigs, L.C., R.L. Beuheim, T.L. Jones (editors) 2000. *Interpretation of Tracer Tests Performed in the Culebra Dolomite at the Wast Isolation Pilot Plant Site*, SAND97-3109

1995-96 Tracer Tests

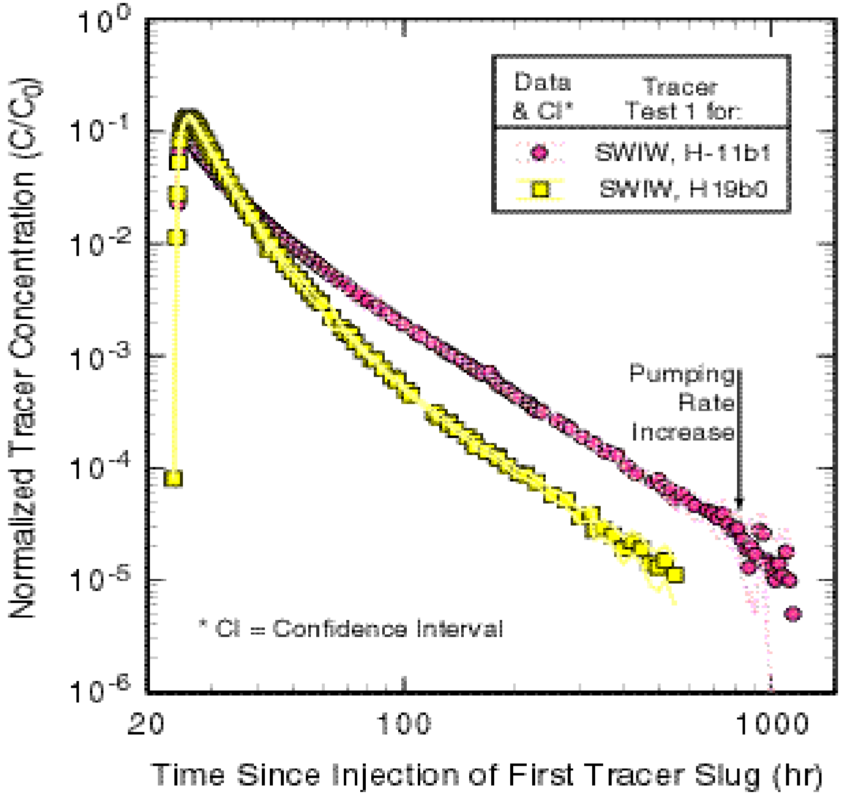
- Performed in fractured Culebra dolomite
- SWIW and convergent-flow tests performed
- Convergent-flow tests involved three and six different flow paths—preliminary testing performed before locations for final three tracer-injection wells determined
- Employed tracers with different diffusion coefficients
- Tracers injected over full and partial thicknesses of Culebra
- Two different pumping rates used
  - Different velocities allow different times for diffusion

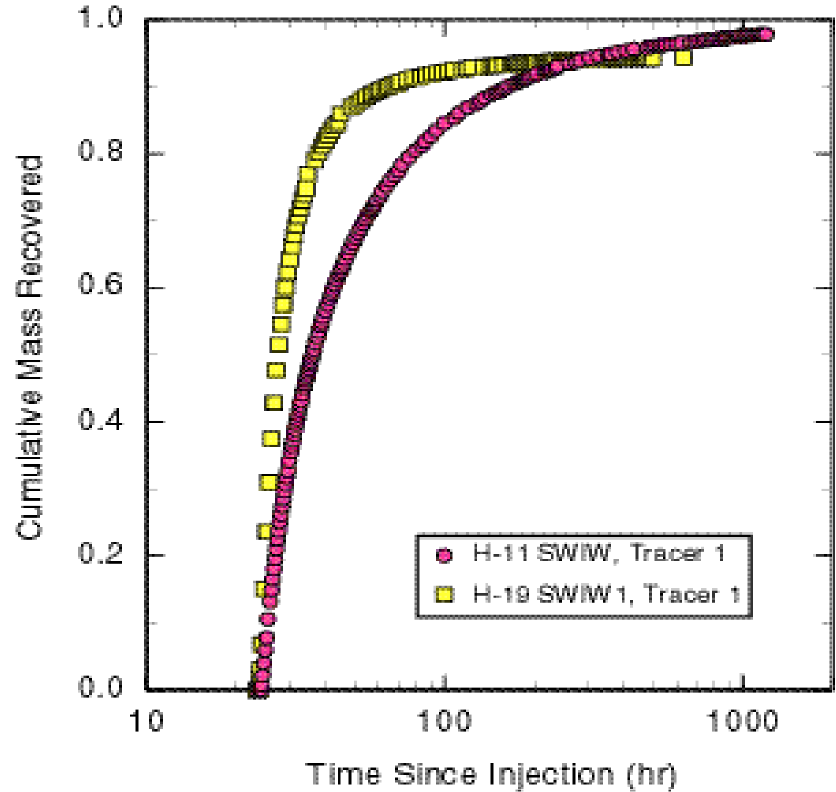
Well Locations for H-19 Tracer Test



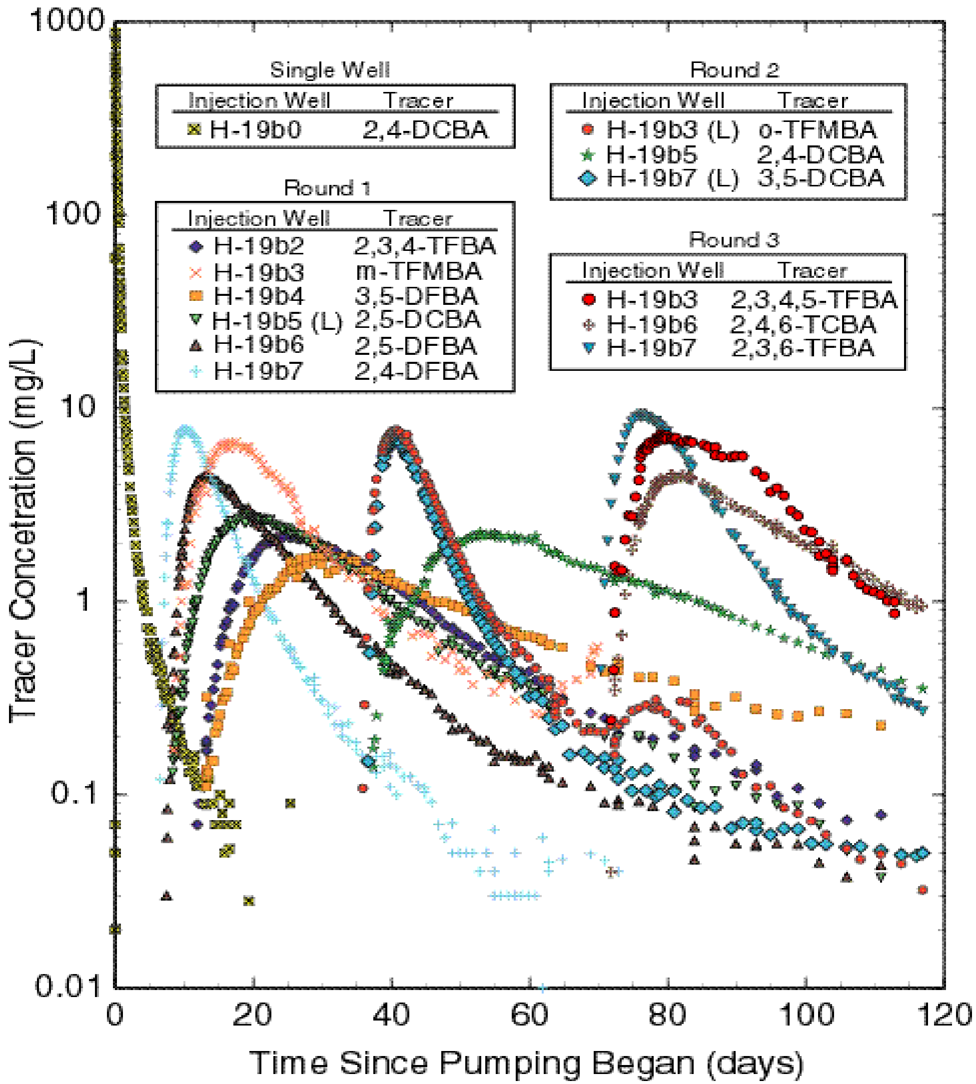
SWIW Test Data

- 94-98% mass recovered





H-19 Convergent-Flow Tracer Test Data



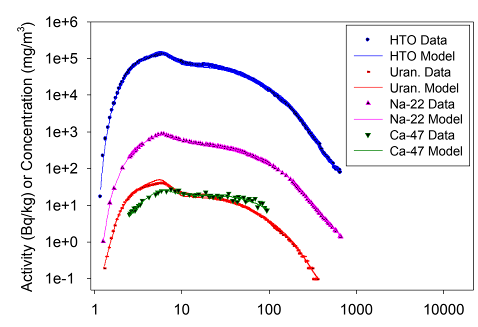
- 74-103% mass recovered for full and lower Culebra intervals; 5-18% mass recovered for upper Culebra intervals

Äspö TRUE: Tracer Retention & Understanding Experiment

- **Convergent Flow Tracer Tests conducted in a fracture zone (Feature A) at the Äspö URL**
  - **10 different tracers (mainly radionuclides including tritium)**
  - **Broad range of sorption strength across tracers**
  - **One pumping well, 2 injection wells**
  - **Different pumping rates**
  - **Multiple rock materials: gauge, mylonite, altered and unaltered diorite, fine-grained granite**
  - **Scales of tests are about 5 meters and 1000 hours**

The Äspö Task Force on Modelling of Groundwater Flow and Transport of Solutes

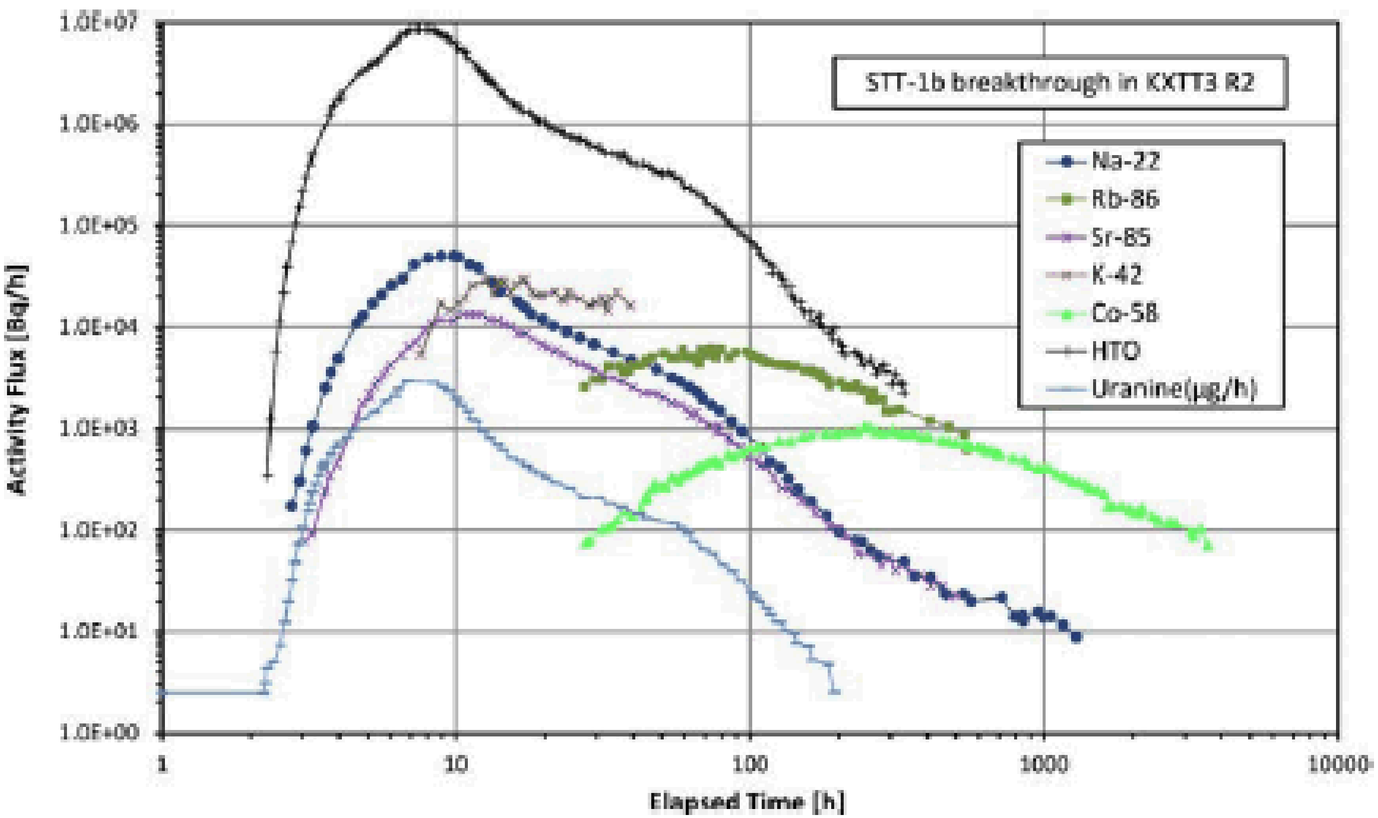
Tracer test program administered by international Äspö Task Force (JNC and Sandia are members)



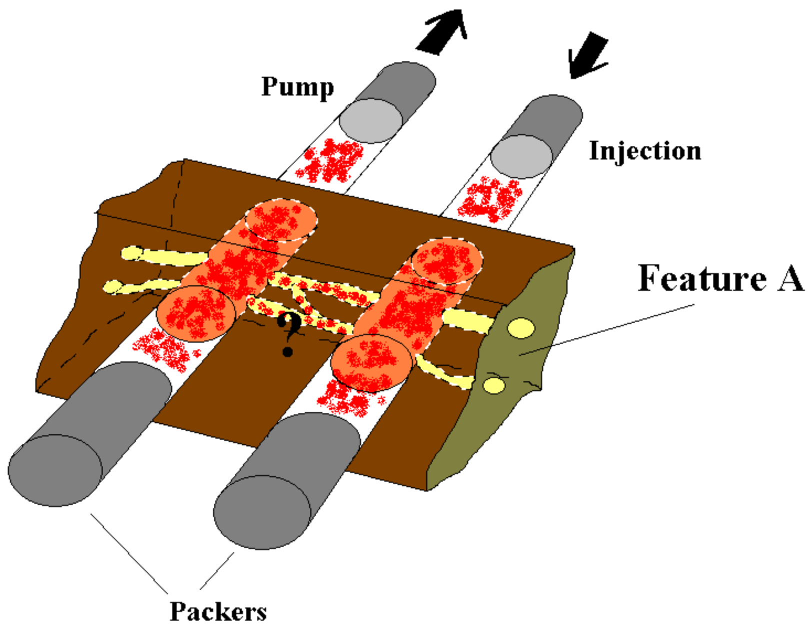
Example fits to data made with SNL multirate model

Äspö STT-1b Tracer Test

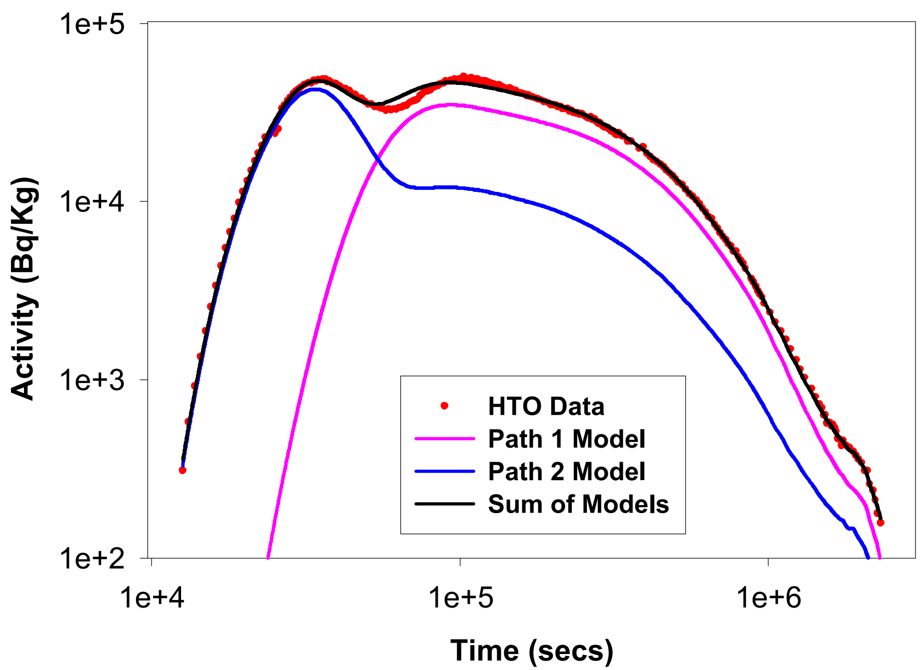
- **Convergent-flow test with 5-m separation**
- **4 nonsorbing tracers (uranine, HTO, ^{82}Br, ^{131}I)**
- **6 sorbing tracers (^{22}Na, ^{42}K, ^{85}Sr, ^{99}Tc, ^{58}Co, ^{86}Rb)**



TRUE-1 Tests at Äspö: STT-2



STT-2 tracer test shows evidence of 2 discrete pathways between wells. Multirate mass-transfer model was adapted to capture this behavior.



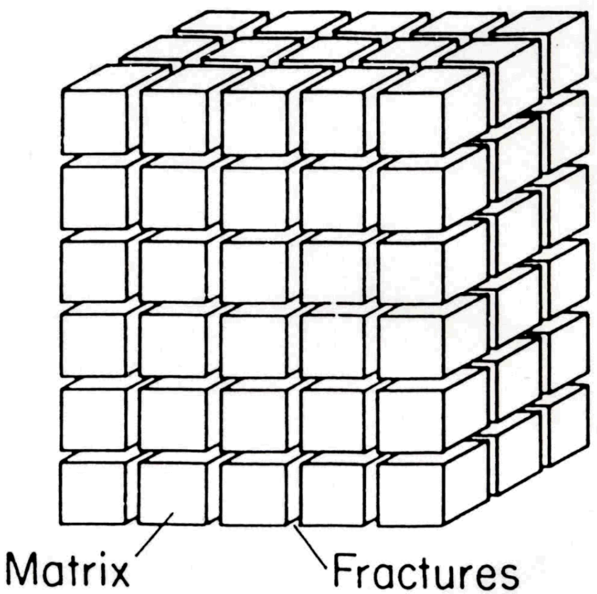
Use inverse model to determine parameters for each pathway and the amount of tracer mass traveling along each pathway

Recommendations for Tracer Testing

- Perform a preliminary test to refine the design of the “real” test
- Combine use of SWIW and convergent-flow tests
  - SWIW tests sensitive to multiple rates of diffusion
  - Convergent-flow tests sensitive to advective porosity
- Vary pumping rates and use multiple tracers with different diffusion coefficients
  - Use to discriminate matrix diffusion from heterogeneity
- “Validate” results by blind prediction of results expected for as-yet-untested flow path(s)
  - Convert tracer-injection well from first test(s) to pumping well for new test, and perform new tests with different orientation of hydraulic gradients

Multi-Rate Transport (STAMMT-L)

Dual-Porosity Model



The classic dual-porosity representation of a fractured medium is the “sugar-cube” model

Make block size large enough to look “infinite” throughout time of tracer test to match a -3/2 slope

To match an observed breakthrough curve that does not have a –3/2 slope, the amount of dispersion and the matrix block size are adjusted

The Real World

Large blocks: bigger capacity, less surface area per aquifer volume, slower diffusion rate

Small blocks: small capacity, more surface area per aquifer volume, faster diffusion rate

Solute accesses all blocks simultaneously

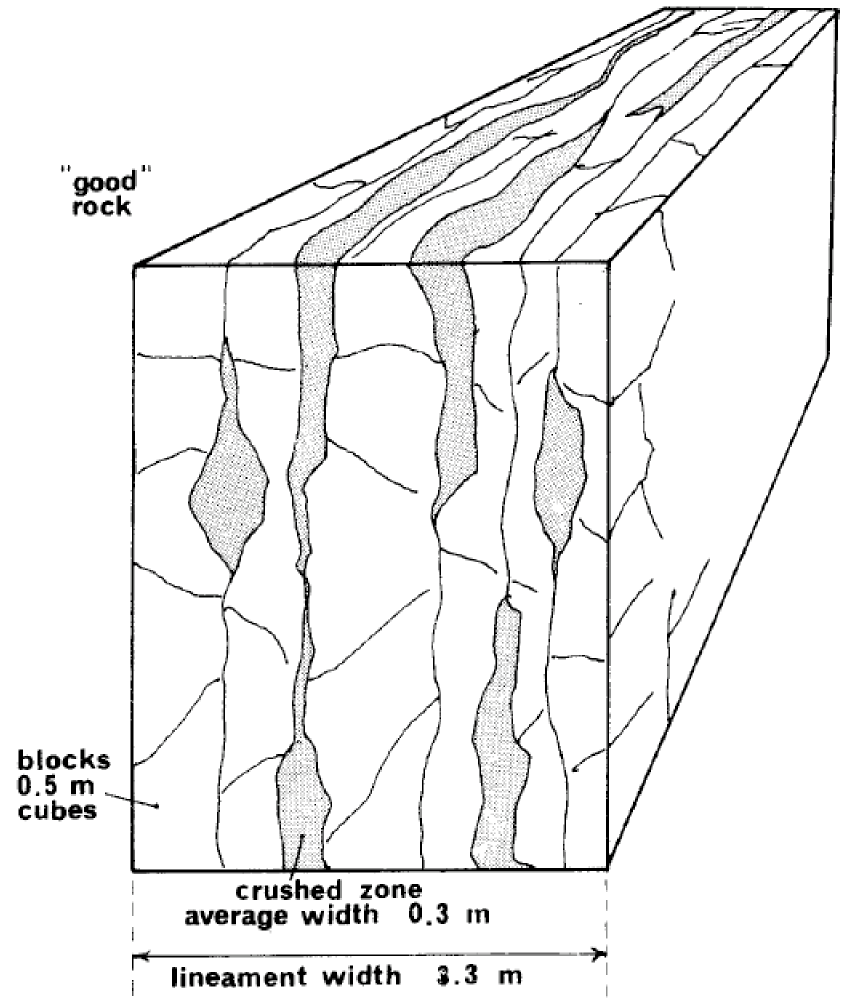
Cemented breccia zone at Yucca Mountain, Nevada

Multirate Conceptualization

Initial models developed for transport along fracture zones in granitic rock.

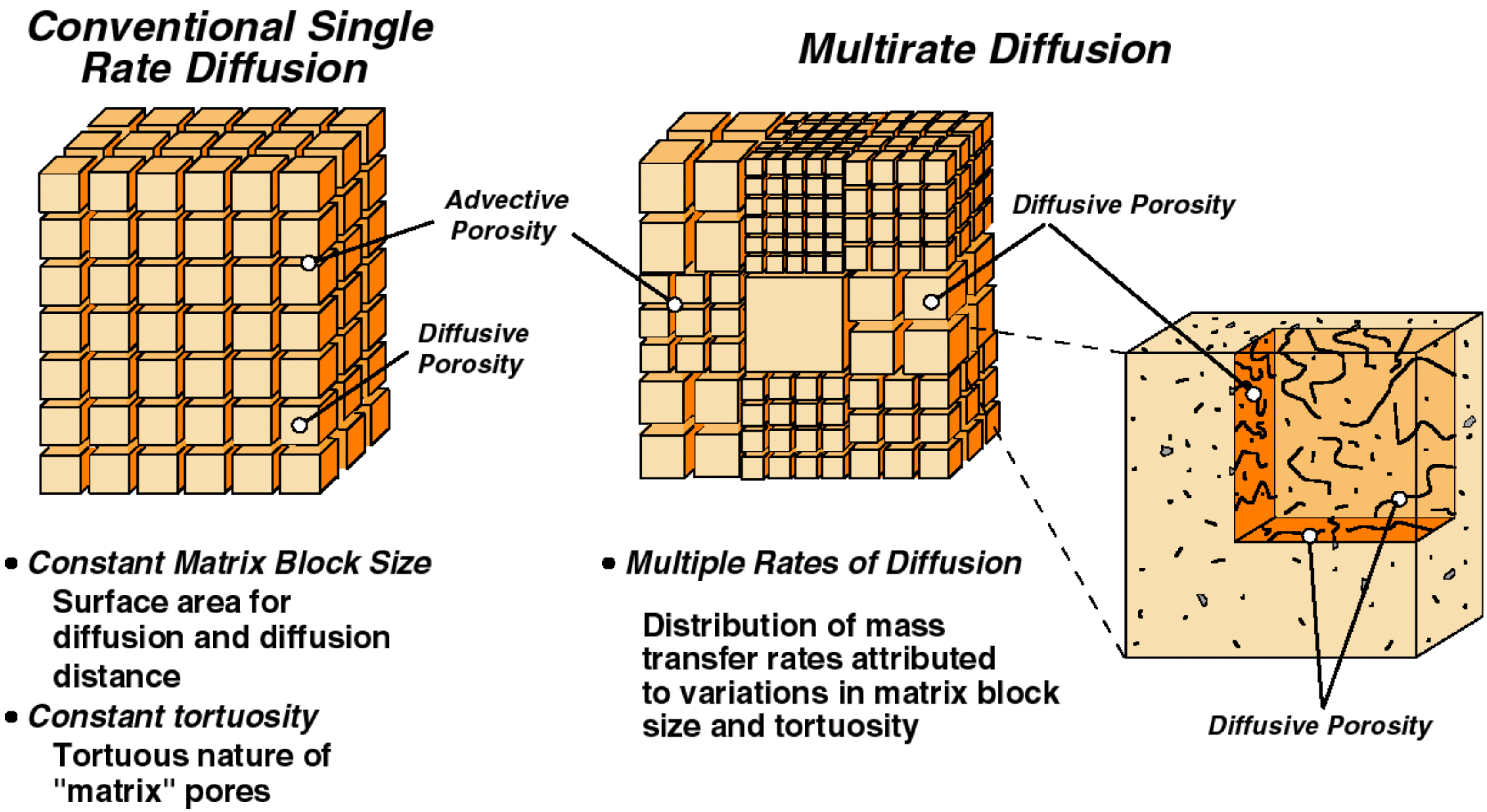
Generally two zones: intact granite and a "crushed" zone that included open fracture, fault gouge, and weathered rock

Important concept that not all block sizes are equal and that the impact of this variable geometry on surface area to volume relationships has significant impact on soprtion and diffusion properties

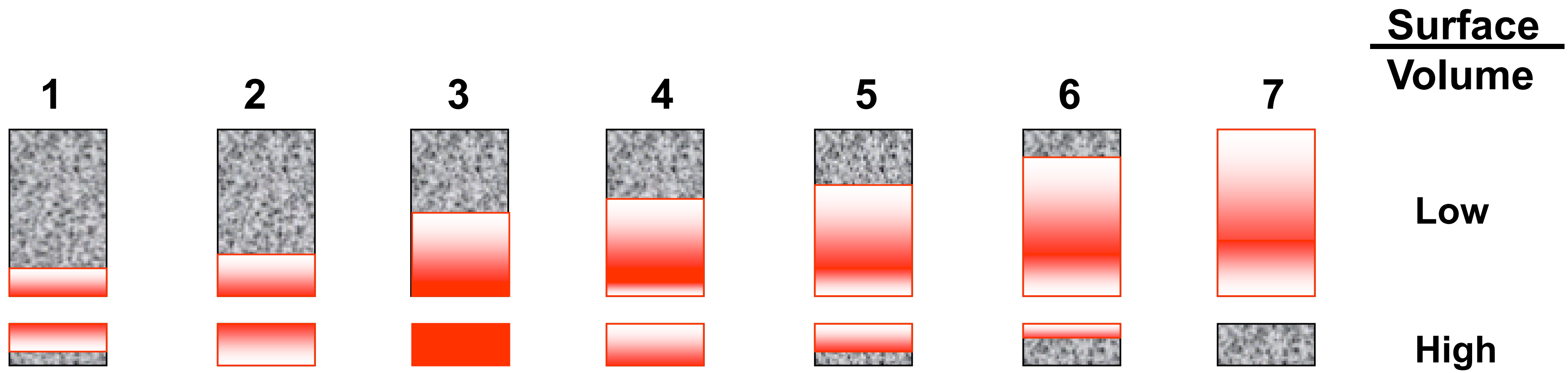


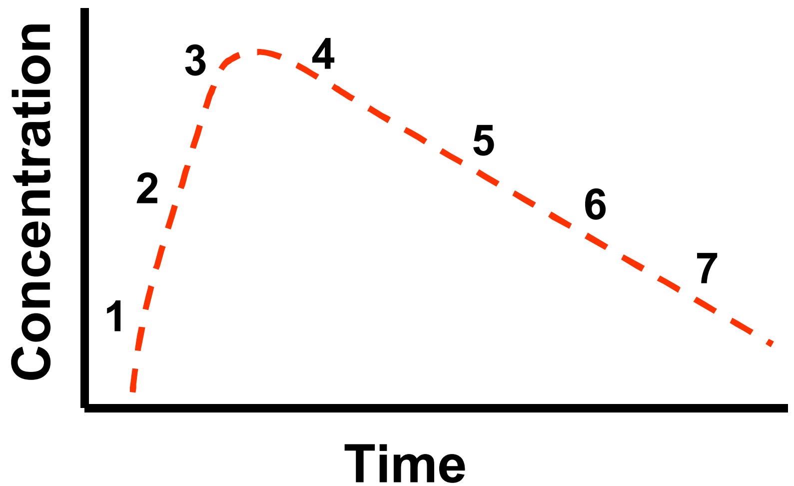
Neretnieks and Rasmuson, 1983, An approach to modling radionuclide migration in a medium with strongly varying velocity and block sizes along the flow path, SKB Report, 83-69

Multi-Porosity Conceptualization



Matrix Block Size





Slower mass transfer from matrix results in shallower (longer) tail

Different rates of mass transfer create different slopes in late time tail

Terminology Details

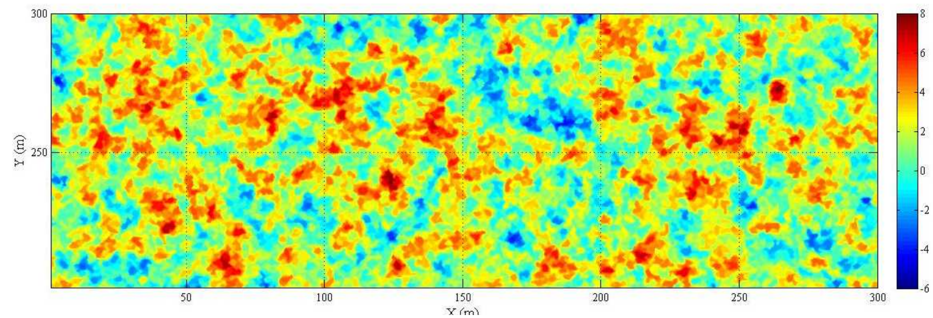
Consider two domains that correspond to the dual-porosity model:

1) Advective (Mobile) Domain

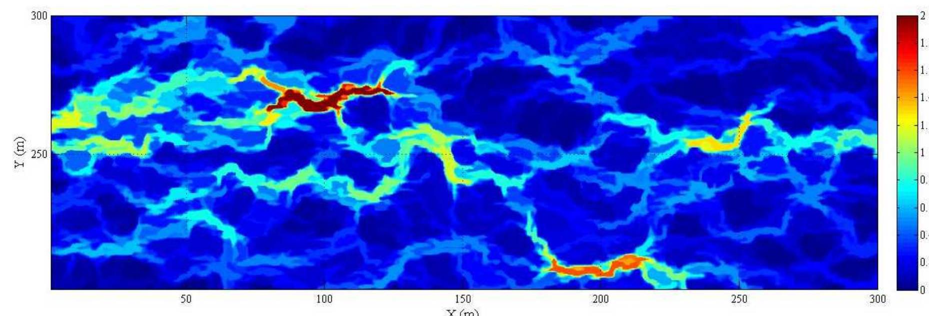
2) Diffusive (Immobile) Domain

Focus here on fractured rock, but concepts also apply to flow in strongly heterogeneous media

SGSIM realization of ln K



Resulting velocity; gradient from left to right



Simulations from Kate Klise

Zinn and Harvey, 2003, When good statistical models of aquifer heterogeneity go bad: A comparison of flow, dispersion, and mass transfer in connected and multivariate Gaussian hydraulic conductivity fields, Water Resources Research, 39 (3)

Transport Equation: Another Look

$$\frac{\partial}{\partial x}\left(D\frac{\partial c}{\partial x}-vc\right)=\frac{\partial c}{\partial t}$$

ADE in general form

$$\frac{1}{R_m}\frac{\partial}{\partial x}\left(D\frac{\partial c}{\partial x}-vc\right)=\frac{\partial c}{\partial t}+\Gamma(x,t)$$

Addition of retardation and a source/sink term for solute mass

c = solute concentration in the advective domain M/L^3

v = average advective velocity (L/t)

$D=\alpha_l v+D^*$ = hydrodynamic dispersion (L^2/t); limit to $a_l v$

R_m = retardation coefficient in the (mobile) advective domain (-)

$\Gamma(x,t)$ = source/sink for mass transfer with diffusive domain (M/L^{3}t)

Expand with Domains

$$\frac{1}{R_m}\frac{\partial}{\partial x}\left(D\frac{\partial c}{\partial x} - vc\right) = \frac{\partial c}{\partial t} + \Gamma(x,t)$$

Rewrite in multirate form with specification of mobile and immobile zone components

$$\frac{\partial}{\partial x}\left(\frac{\alpha_l\,|v|}{R_m}\frac{\partial c_m}{\partial x}\right) - \frac{v}{R_m}\frac{\partial c_m}{\partial x} = \frac{\partial c_m}{\partial t} + \int_0^{\infty} b(\alpha_{im})\frac{\partial \hat{c}_{im}}{\partial t}\,d\alpha_{im}$$

Hydrodynamic dispersion

Multirate exchange with immobile zone

Multirate exchange is defined by two terms:
α mass-transfer rate
b capacity associated with each rate

Haggerty, et al., Water Resources Research, 2001
McKenna, et al., Water Resources Research, 2001

Lognormal Distribution of Rates

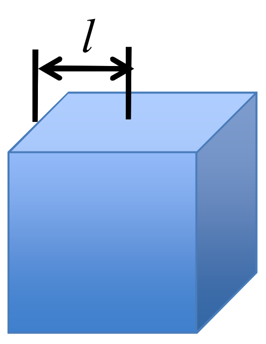
$$b(\alpha_{im}) = \frac{\beta_{tot}}{\sqrt{2\pi}\sigma_{im}\alpha_{im}} \exp\left\{-\frac{[\ln(\alpha_{im}) - \mu_{im}]^2}{2\sigma_{im}^2}\right\}$$

$$\alpha_{im} = \frac{D_\alpha}{l^2} = \frac{D_{aq}\tau}{l^2 R_{im}}$$

$$D_\alpha = D_{aq}\tau$$

$$\beta_{tot} = \frac{\phi_{im} R_{im}}{\phi_m R_m}$$

The total capacitance of the immobile zone, β_{tot}, is equal to the ratio of solute in the immobile zone to mobile zone at equilibrium conditions (generally >> 1)



The matrix block size, *l*, is the distance from the mobile/immobile zone interface to the center of the immobile zone

Multirate Mass Transfer

$$\frac{\partial}{\partial x}\left(\frac{\alpha_l |v|}{R_m}\frac{\partial c_m}{\partial x}\right) - \frac{v}{R_m}\frac{\partial c_m}{\partial x} = \frac{\partial c_m}{\partial t} + \int_0^\infty b(\alpha_{im})\frac{\partial \hat{c}_{im}}{\partial t}\,d\alpha_{im}$$

mobile domain

$$\frac{\partial c_{im}}{\partial t} = D\frac{\partial^2 c_{im}}{\partial z^2}$$

immobile domain

Initial conditions:

$$c(x,t=0) = c_{im}(x,z,t=0) = c_0$$

Uniform concentration at start

Boundary conditions:

$$c_{im}(\alpha_{im}, z=l) = c_m \qquad 0 < \alpha_{im} < \infty$$

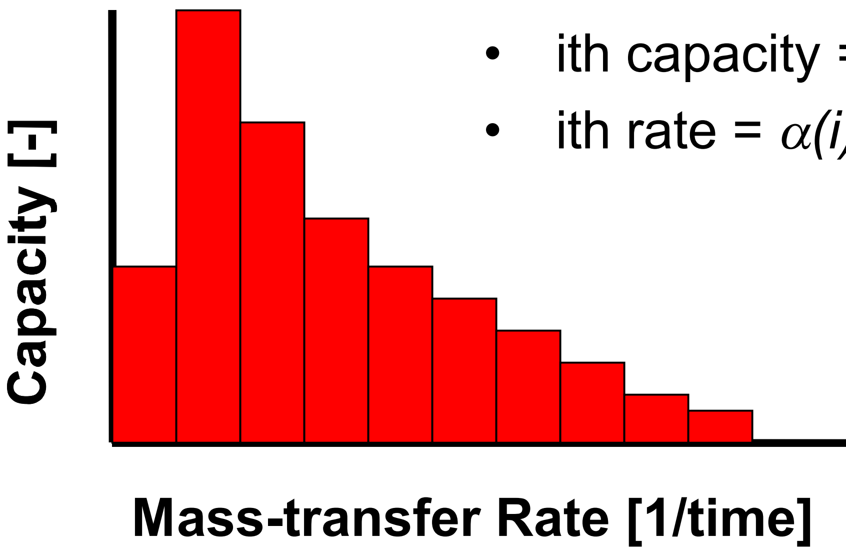
Concentration is the same in the two zones at the mobile/immobile zone interface

$$\frac{\partial c_{im}}{\partial z}(\alpha_{im}, z=0) \qquad 0 < \alpha_{im} < \infty$$

Concentration gradient at the center of the block is zero

Multirate Distribution

Parameterize the capacity of the matrix to uptake solute with a <u>log-normal distribution</u> of mass transfer rate coefficients



- ith capacity = *b(i)*
- ith rate = $\alpha(i)$

$$\beta_{tot} = \sum_{i=1}^{N} b(i) = \frac{\phi_{im} R_{im}}{\phi_m R_m}$$

$$\alpha_i = \frac{D_{aq}\tau}{l^2 R_{im}}$$

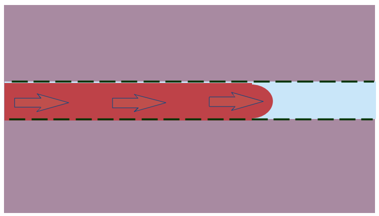
Log-normal distribution of rate coefficients is defined by mean (μ) and standard deviation (σ)

$$\mu = \frac{1}{N}\sum_{i=1}^{N}\log(\alpha(i)) \qquad \sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(\log(\alpha(i)) - \mu)}$$

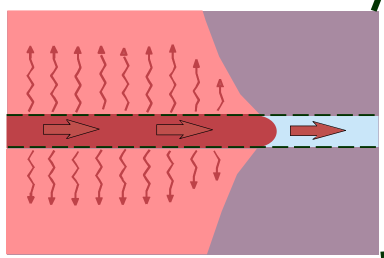
Analysis of Tracer Tests to Provide a Defensible Model for WIPP PA

Conceptual Model

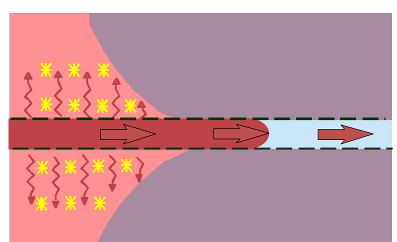
Single-Porosity Fracture-Only Transport



Double-Porosity Nonreactive Transport (Physical Retardation)

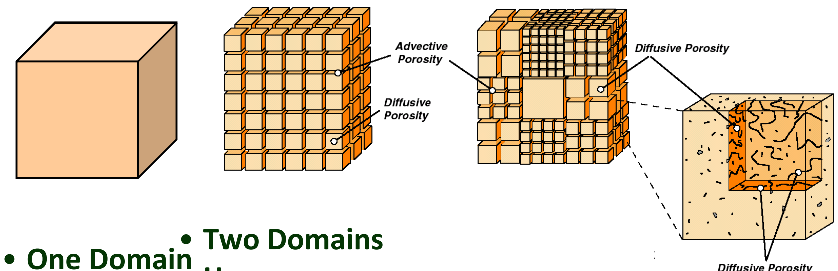


Double-Porosity Reactive Transport (Physical and Chemical Retardation)



Numerical Implementation

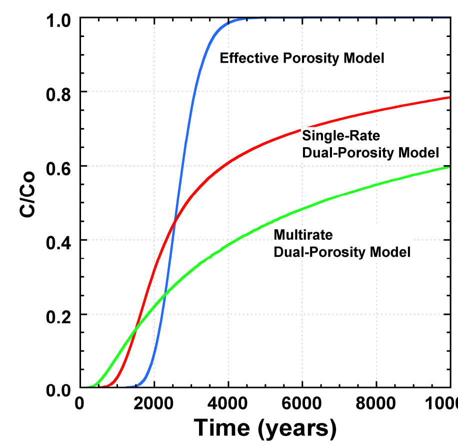
Effective-Porosity Model | **Conventional Single-Rate Diffusion** | **Multirate Diffusion**



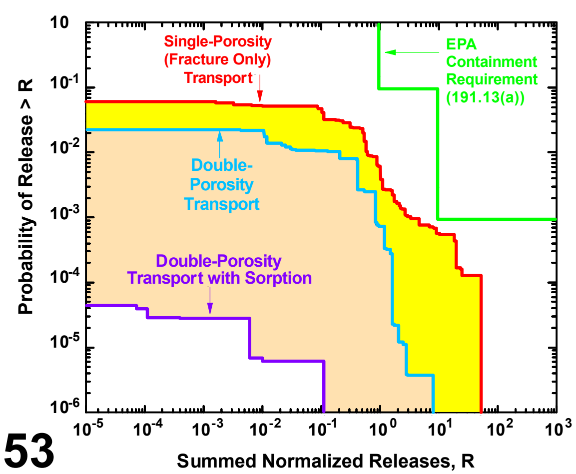
- One Domain

- Two Domains
- Homogeneous Matrix and Fracture

- Two Domains
- Heterogeneous Matrix and Fracture



Comparison of mechanisms and WIPP release probability



Integration of Tracer Tests into Site Licensing

Tracer Tests: 1980-1988

- 5 locations
- Types of tests
  1) Convergent-flow tests
  2) Two-well recirculating tests
- Analysis method:
  1) radial (1D) single-rate, double-porosity model

Recognized need to reduce conceptual model and data uncertainty (1994)

Tracer Tests: 1995-1996

- 2 locations
- Types of tests:
  1) Convergent-flow tests
  2) Single-well injection-withdrawal tests
- Analysis methods:
  1) 2D (heterogeneous) single-rate, double-porosity model
  2) radial (1D) single-rate, double-porosity model
  3) radial (1D) multirate, double-porosity model

Comments of Independent Reviewers

- Questioned matrix diffusion as mechanism for retardation
- Suggested alternative mechanisms:
  1) Channeling caused heterogeneity
  2) Delayed release of tracer from the injection wells

Use for Compliance and Certification

- Confirmed matrix diffusion as a mechanism for retardation
- Provided credible, defensible and realistic model
- Model reviewed and accepted by EPA-mandated Conceptual Model and Natural Barriers Peer Review Panels
- Provided basis for simplified PA model
- Provided important physical transport parameters for PA
- Provided rationale for parameters