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Transport in Fractured Rock

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Outline

- Fracture transport processes (from perspective of tracer testing)
- Three conceptual models
  - Single Porosity
  - Double Porosity
  - Multi-porosity (multirate)
- Field Examples
  - Aspo TRUE tracer tests
  - Imaging fracture filling

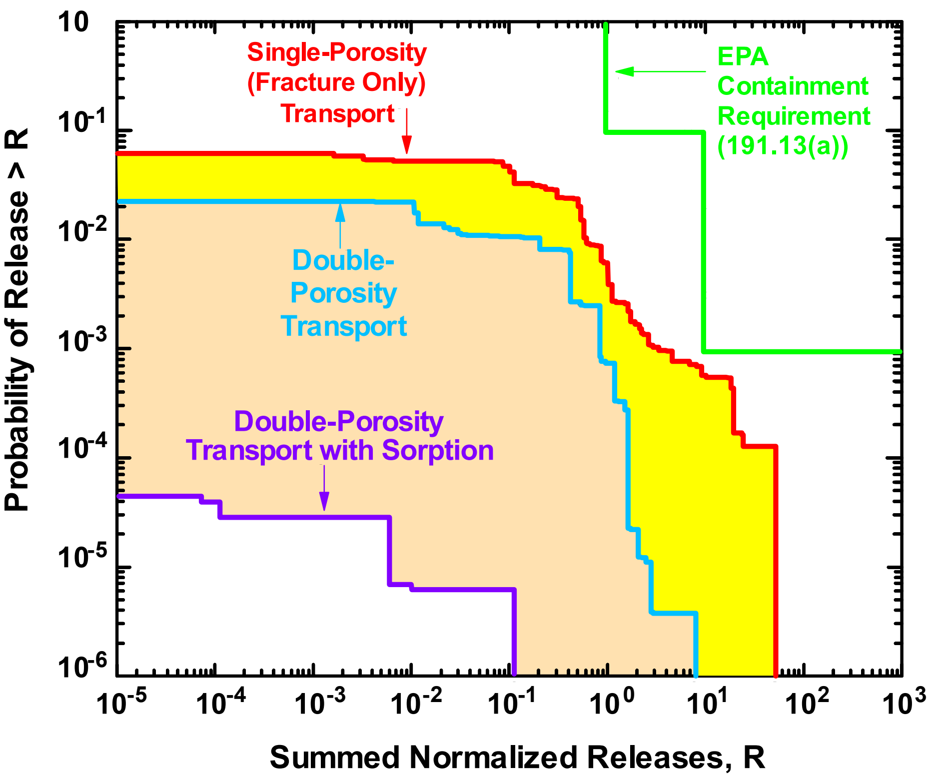
Motivation: WIPP Example

Probabilistic Systems Analysis is used to predict the future performance of the repository – incorporate uncertainty in multiple processes and parameters

Regulations are written to incorporate uncertainty

Mass transfer processes make a <u>huge difference</u> in predicted repository performance

Other situations: hydrothermal mineralization, partitioning tracer tests for DNAPL, groundwater-surface water coupling with bank storage, etc.

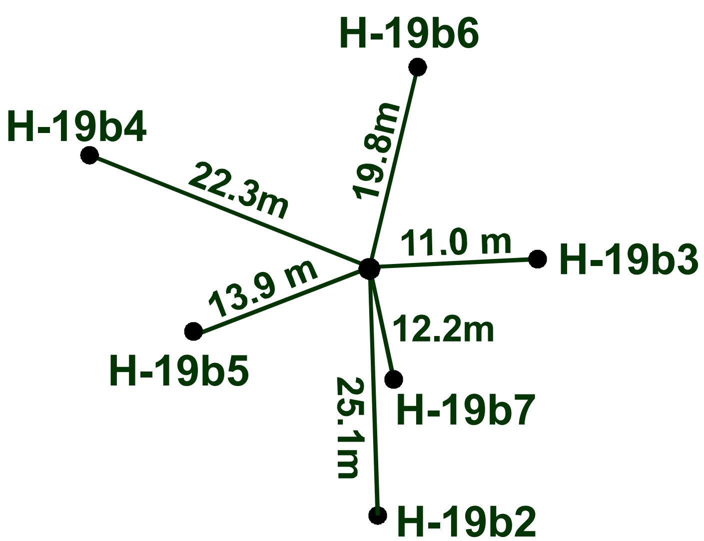


Tracer Testing

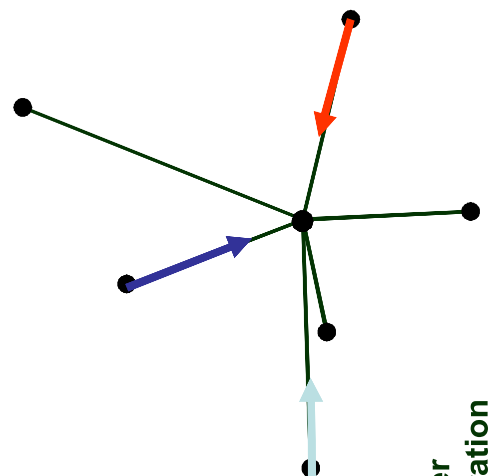
- Inject a solute into the aquifer that does not occur locally in significant amounts
  - Naturally Occurring: Iodine, Chlorine, Deuterium
  - Synthetic: Radionuclides, Chloro/Flourobenzoic acids, polystyrene spheres
- Extract that solute in a pumping well
- Examine the concentration as a function of time (breakthrough curve) to gain insight into aquifer properties
  - Dispersion, porosity, diffusion, sorption

Tracer Testing

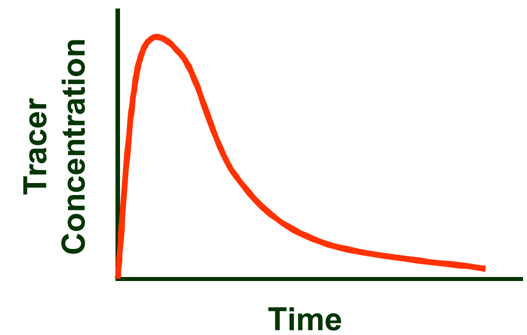
Convergent Flow Tests



Central pumping well with tracer injections at surrounding wells

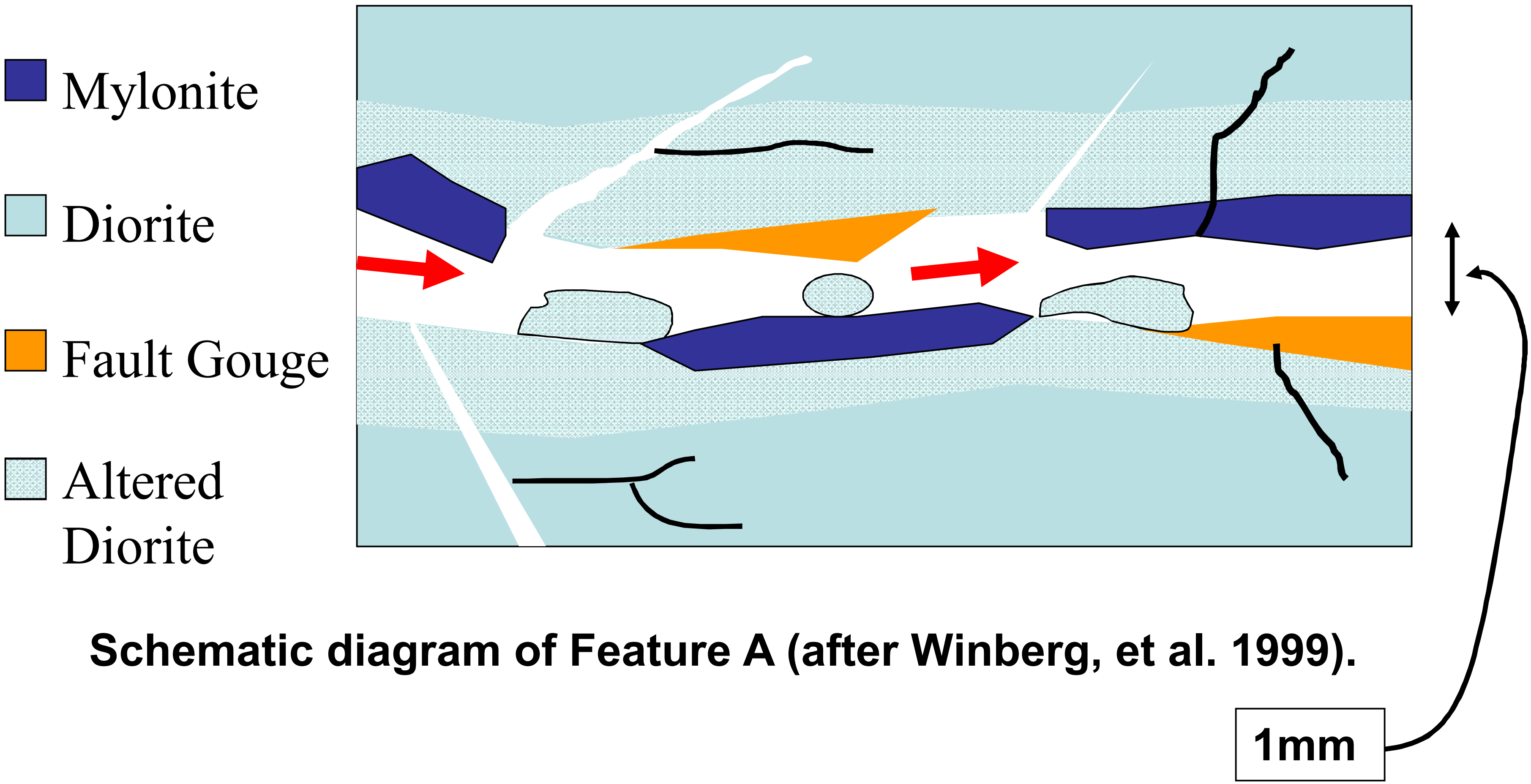


Breakthrough Curve



Injection phase of Culebra H-19 hydropad tracer testing at WIPP Site, SE New Mexico

Conceptual Model: Fractured Granite



Schematic diagram of Feature A (after Winberg, et al. 1999).

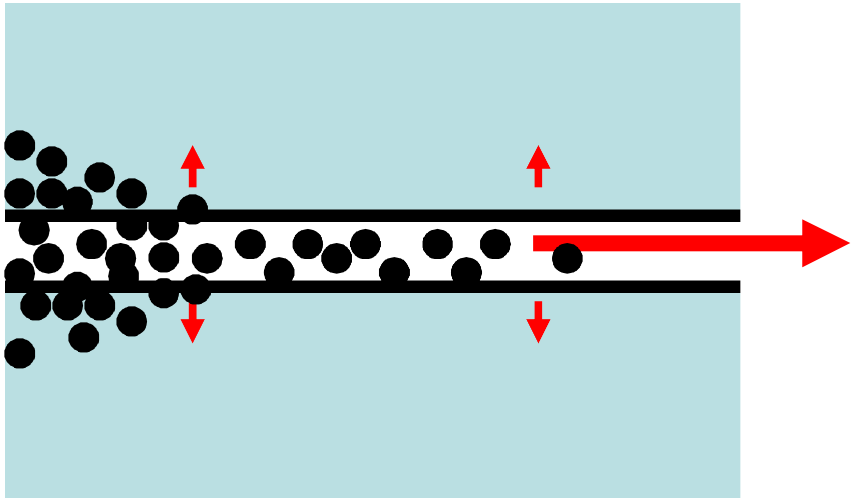
Transport Processes

- **Processes acting in a single fracture:**
  - Dispersion (velocity variation along different flowpaths due to physical heterogeneity in fracture)
  - Diffusion (movement of solute due to concentration gradient – from fracture to matrix and back)
  - Sorption (attachment of solute to fracture walls and matrix pore spaces)

Conceptually, there are two domains in the rock:

1) ***Advective, mobile, fracture***

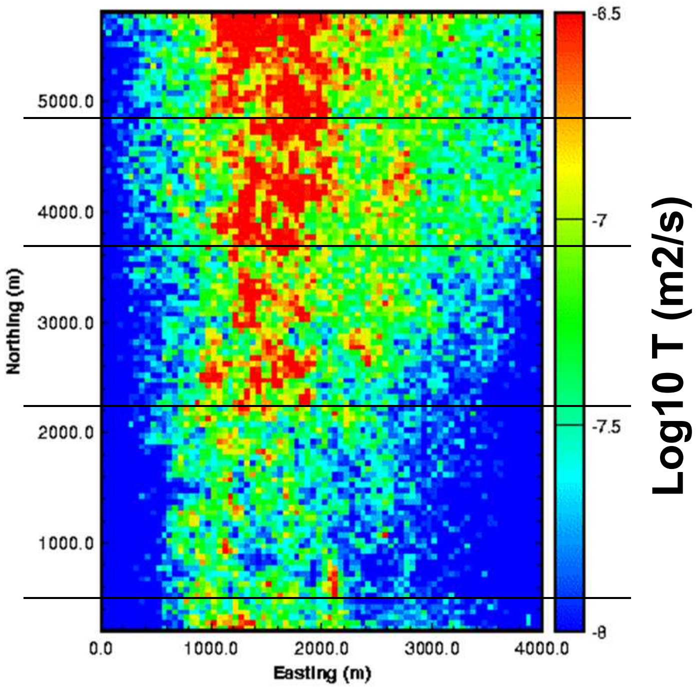
2) ***Diffusive, stagnant, matrix***

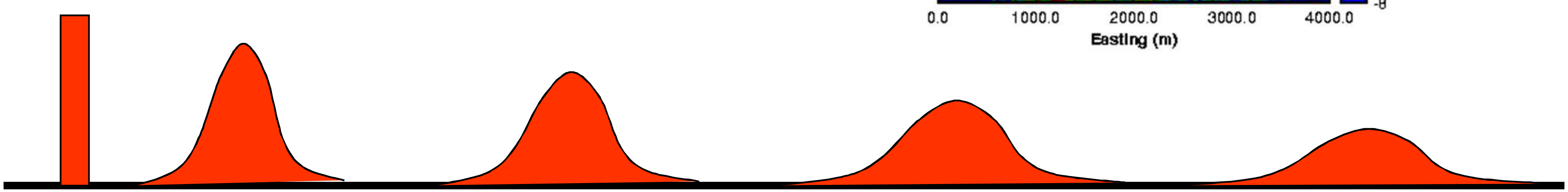


Dispersion

- **Physical process of solute moving at different velocities along different flowpaths**

Flowpaths in a heterogeneous fracture plane





Start **Finish**

Analogy: a 10k running race

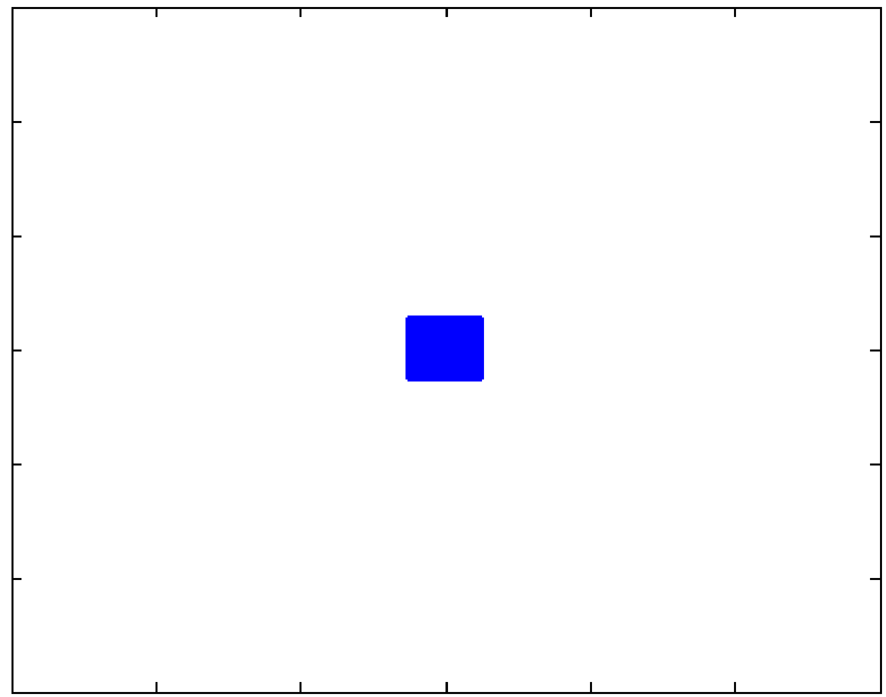
Diffusion

- **Spreading of a solute due to a concentration gradient**

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

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

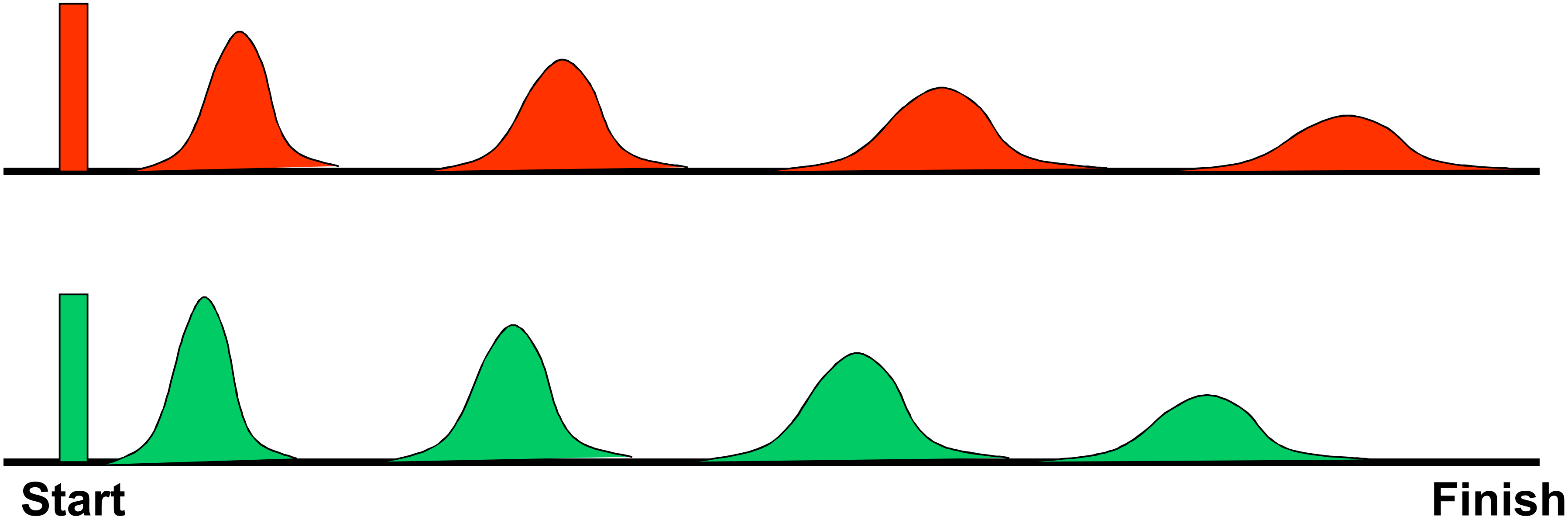
D **is a function of the material(s) through which the solute is diffusing**



Analogy: Drop of dye in an aquarium

Sorption

- **Adherence of solute onto a surface (fracture or pore)**

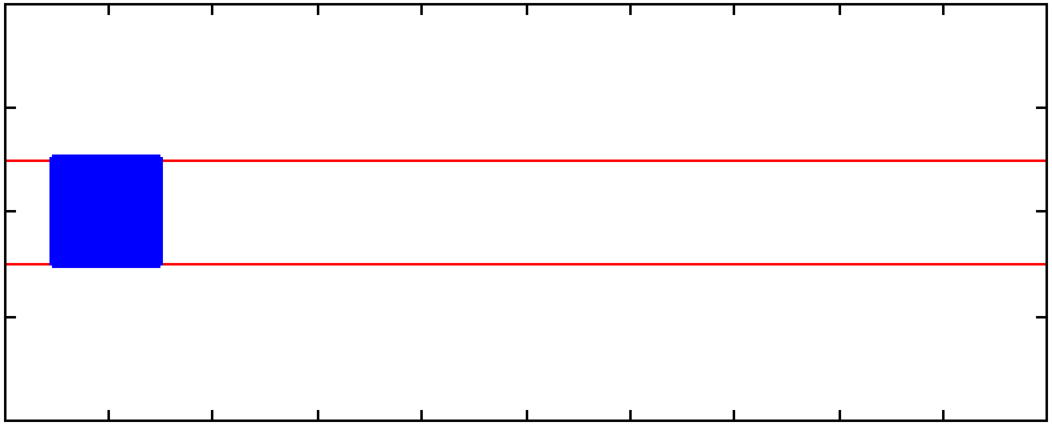


Analogy: A 10k running race with faulty shoe laces

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

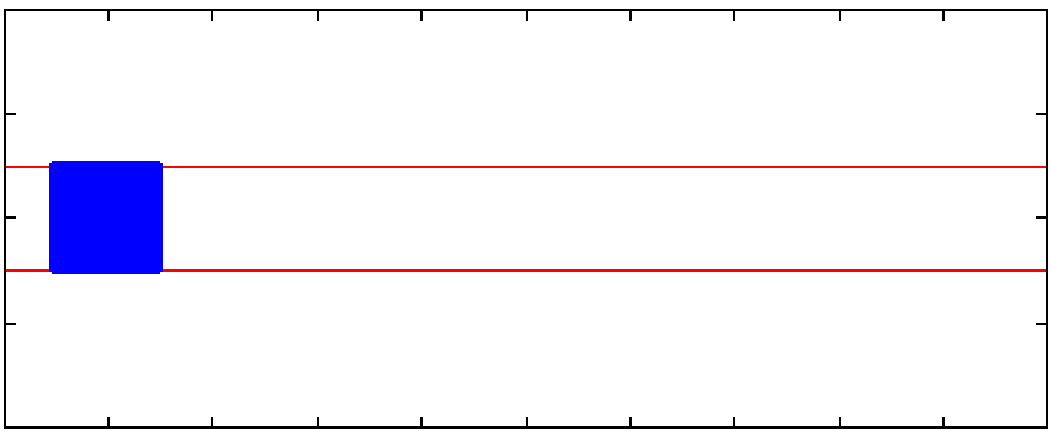


Also referred to as "single-porosity" transport

Dispersion and Diffusion

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

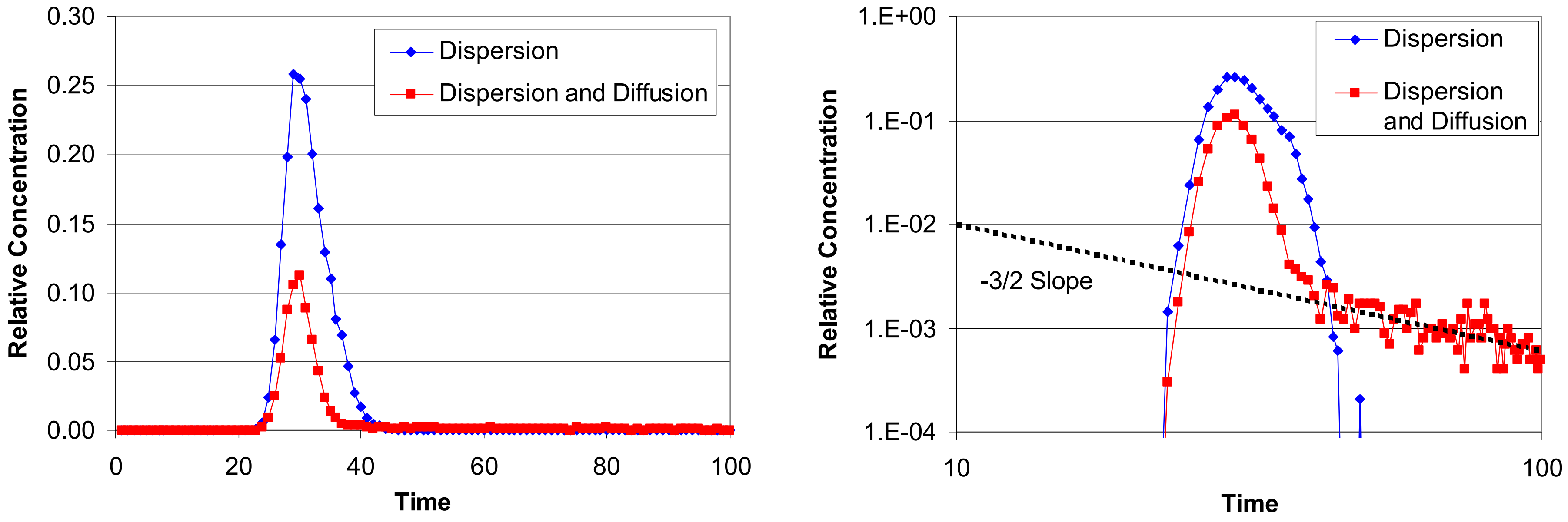
Hydrodynamic dispersion and mass-transfer are active



Also referred to as "dual-porosity" transport

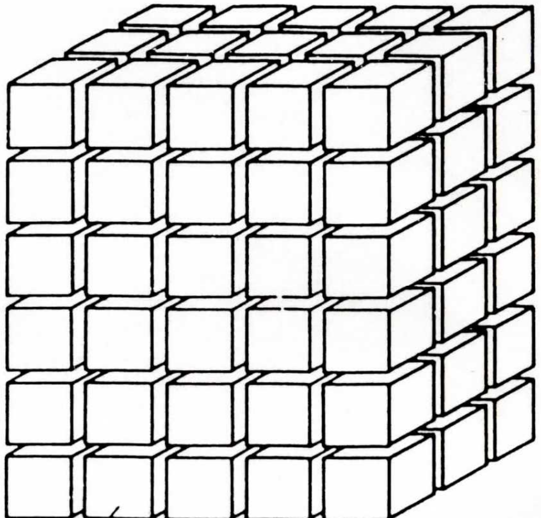
Breakthrough Curves

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, examine results in log-log space***
- ***-3/2 slope is characteristic of diffusion into an infinite medium***

Traditional Dual Porosity Model



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

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

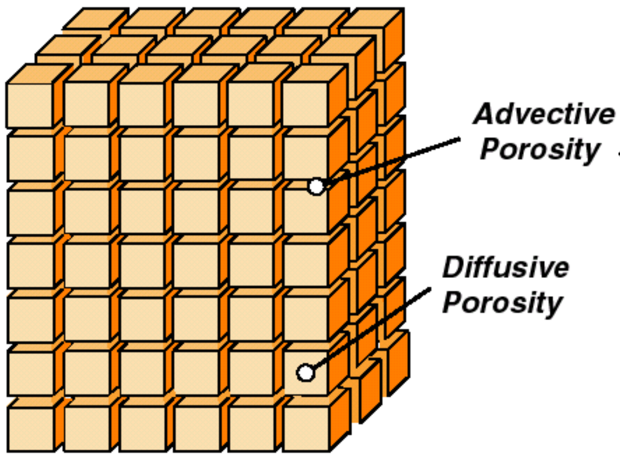
Small blocks: small capacity, more surface area per aquifer volume, faster diffusion rate

Solute accesses all blocks simultaneously

Cemented breccia zone at Yucca Mountain, Nevada

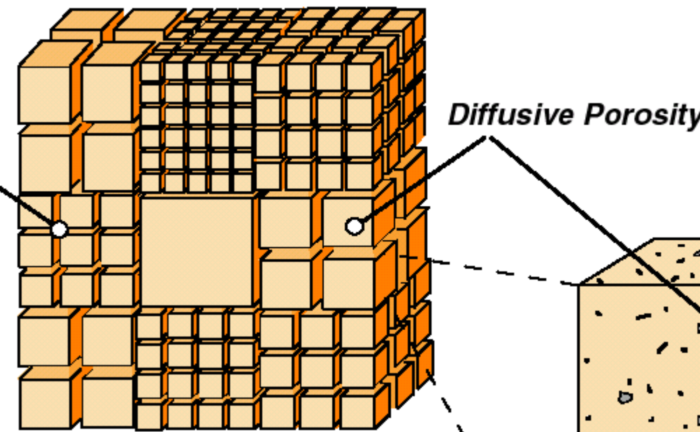
Multi-Porosity Model

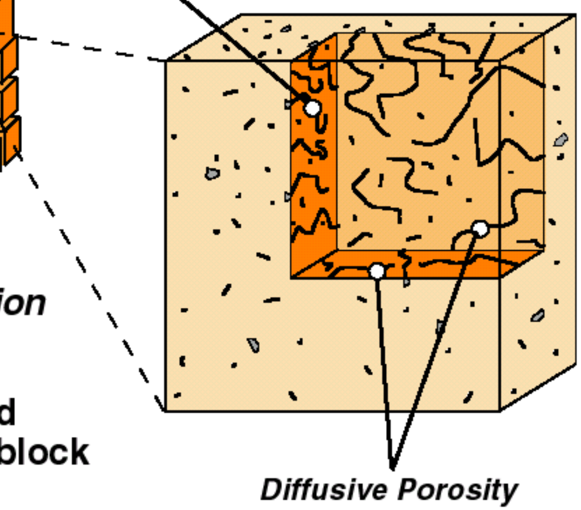
Conventional Single Rate Diffusion



- ***Constant Matrix Block Size***
  Surface area for diffusion and diffusion distance
- ***Constant tortuosity***
  Tortuous nature of "matrix" pores

Multirate Diffusion

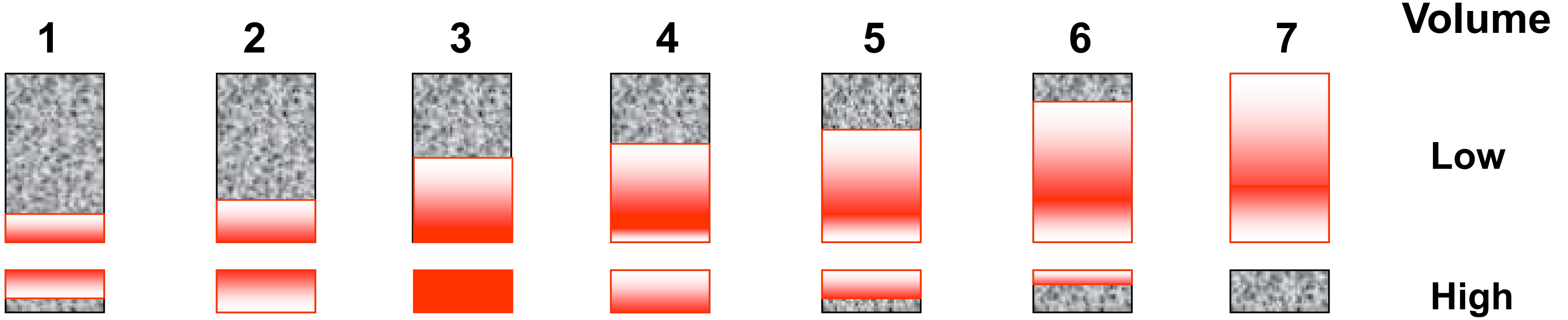


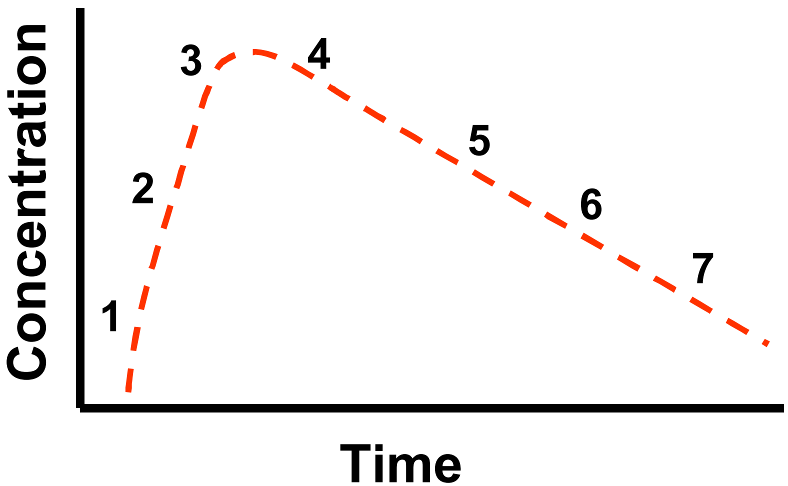


- ***Multiple Rates of Diffusion***

  Distribution of mass transfer rates attributed to variations in matrix block size and tortuosity

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

Transport Equation

Solute transport equation for advection and dispersion in one dimension (x direction) and allowing for mass-transfer through diffusion and/or sorption:

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

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

v = average advective velocity (L/t)

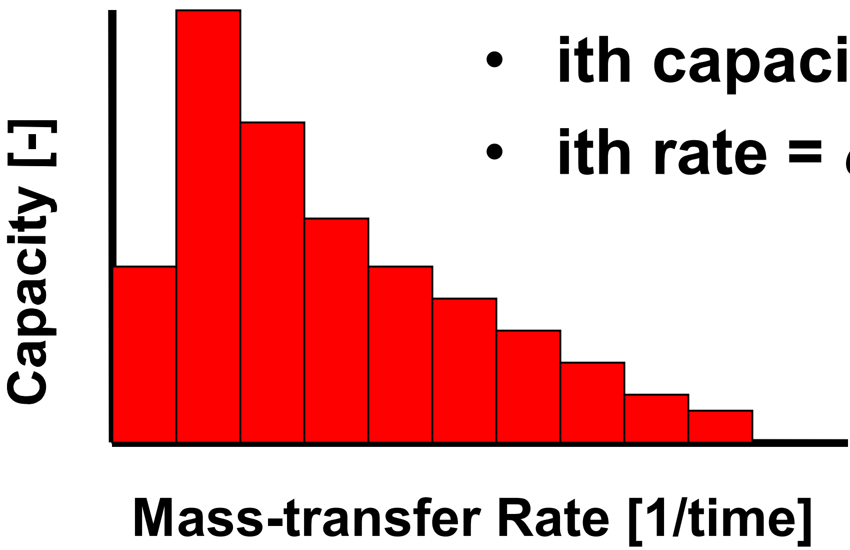
$D = \alpha_l v + D^*$ = hydrodynamic dispersion (L^2/t)

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

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

Model Parameters (mass-transfer)

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 = *α(i)***

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

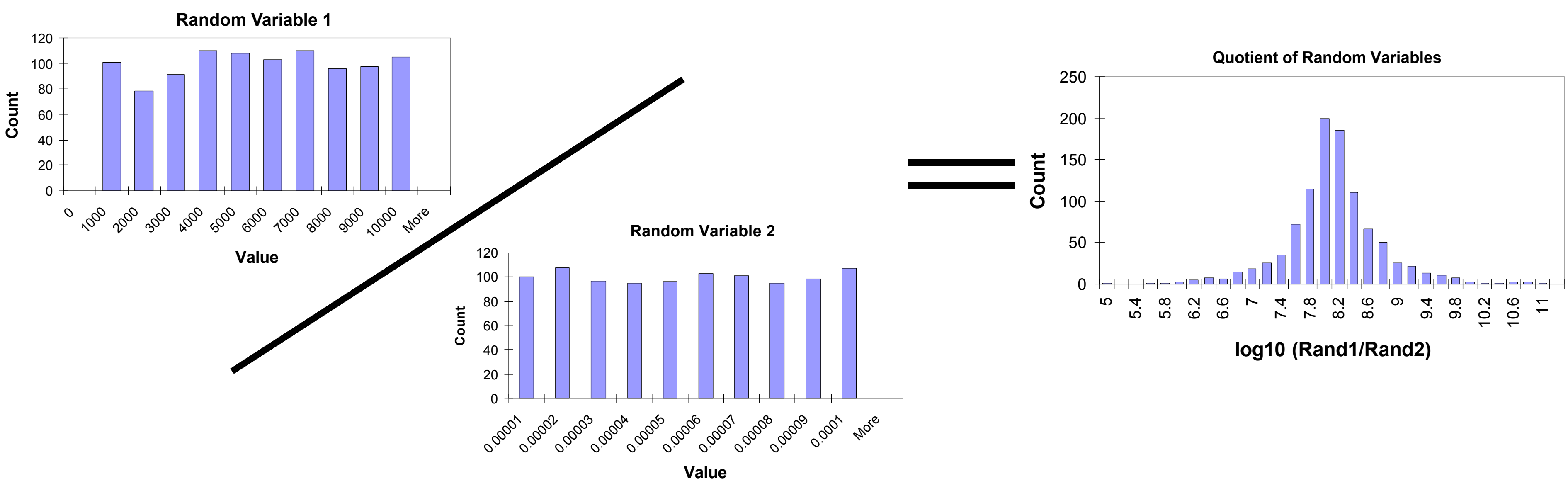
$$\alpha_i = \frac{D_{aq}\tau}{l^2 R_{im}}$$

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

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

Log-Normal Distribution?

Products and Quotients of random variables are log-normally distributed



Random variables: fracture spacing, matrix block size, porosity, diffusion coefficient, tortuosity, sorption site distribution, etc.

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

Model Parameters (sorption)

Mobile zone retardation factor:

$$R_m = 1 + \frac{\rho_b K_{d,m} f_m}{\phi_m}$$

Immobile zone retardation factor:

$$R_{im} = 1 + \frac{\rho_b K_{d,im} f_{im}}{\phi_{im}}$$

ρ_b= bulk density (M/L3)

ϕ_m = mobile zone porosity (-)

ϕ_{im} = immobile zone porosity (-)

K_d = distribution coefficient (L^3/M)

f_m = mass fraction of sorbed phase in equilibrium with mobile zone (-)

f_{im} = mass fraction of sorbed phase in equilibrium with immobile zone (-)

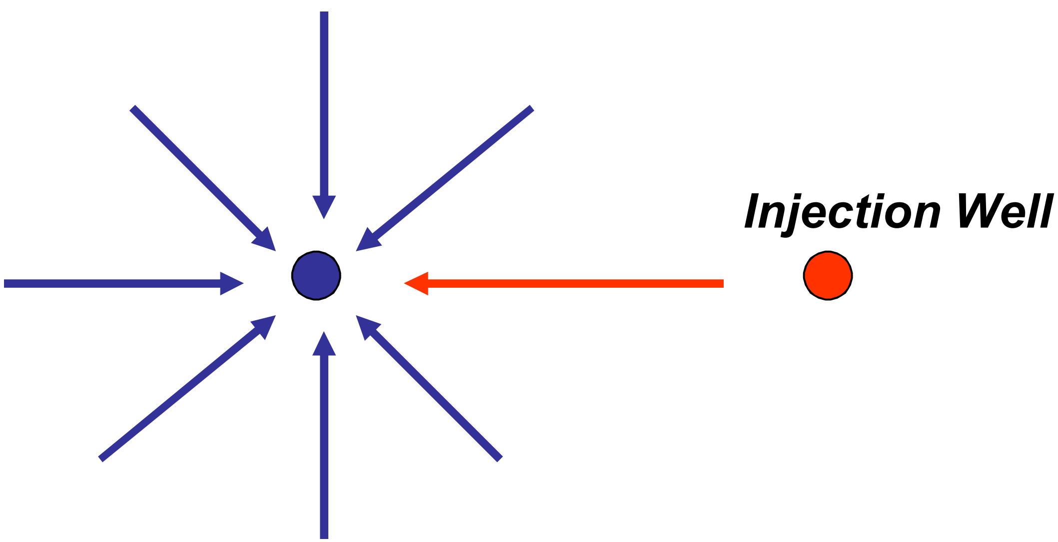
$f_m + f_{im} = 1.0$

Model Parameters (advective)

V_x = 1-D velocity along transport pathway

α_L = longitudinal dispersivity

dilute = dilution factor (not all water coming out of the pumping well has tracer)



$$dilute = \frac{Q_{pump}}{Q_{inject}}$$

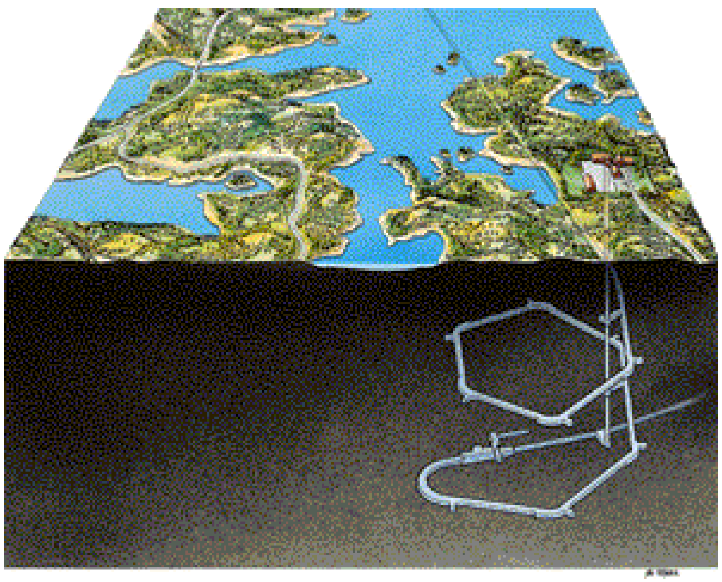
$$dilute = \frac{Q_{pump}}{Q_{inject}} \cdot \frac{1}{RMF}$$

Model Parameters

- **Advective**
  - **Velocity (V_x)**
  - **Dilution Factor (dilute)**
  - **Dispersivity (α_L)**
- **Mass-Transfer**
  - **Total Capacity (β_{tot})**
  - **Mean of lognormal distribution of rates (μ)**
  - **Standard deviation of lognormal distribution of rates (σ)**
  - **Mobile zone retardation factor (R_m)**

Application to Granitic Rocks

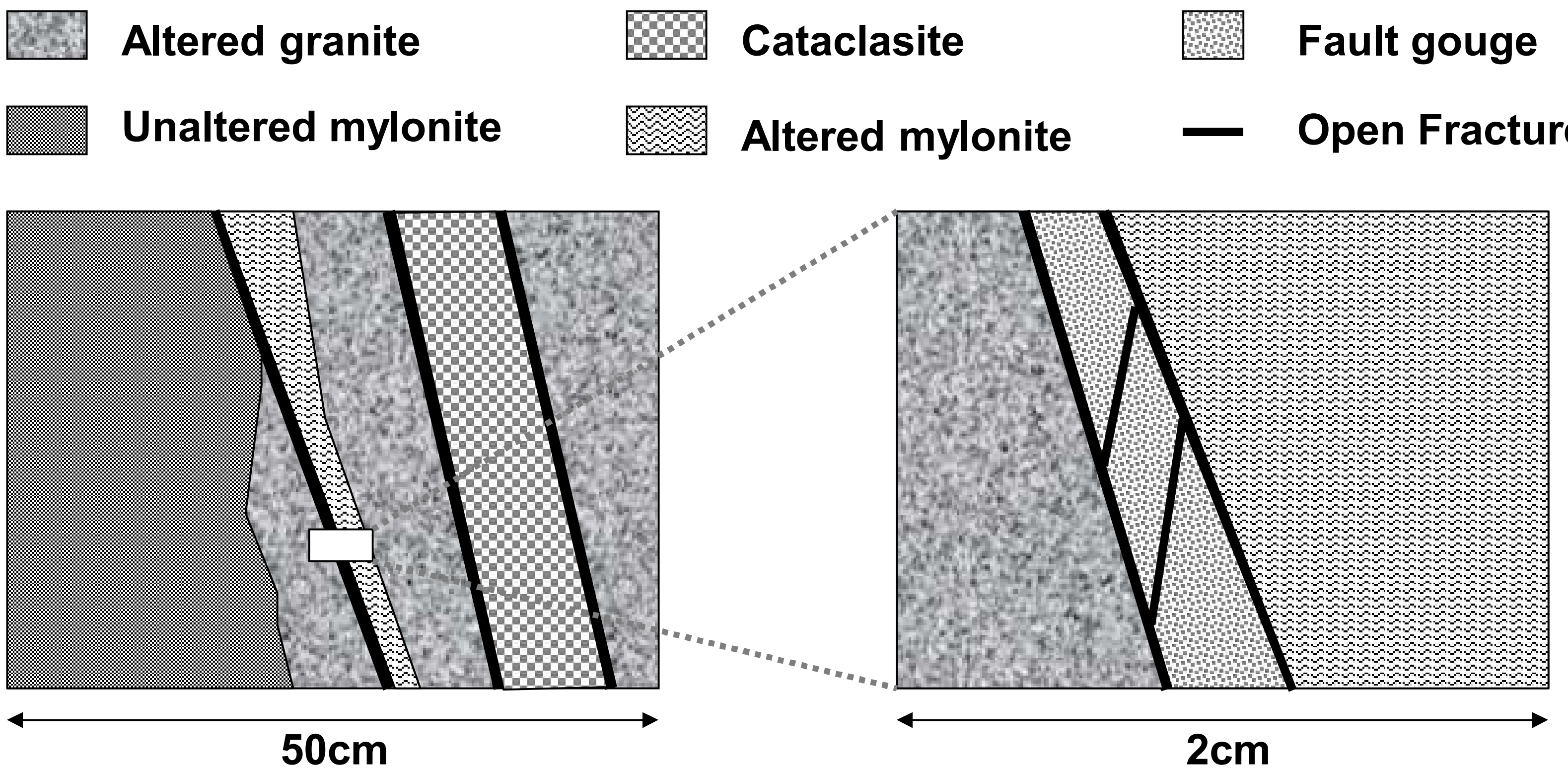
- **Aspo Task Force**
  - 10 nuclear waste organizations from 8 countries
  - Tracer experiments conducted at Aspo underground research laboratory in Sweden



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

Conceptual Model: Feature A

- **Precambrian granites experienced episodic ductile and brittle deformation with hydrothermal mineralization**



After Mazurek, et al., 2003, Jour. Contaminant Hydrology

Experimental Setup

- Tracer testing in a single fracture zone ("Feature A")
- Convergent flow system to a pumping well
- Injection well is 5 m from pumping well
- Multiple radionuclides with different levels of sorption
  - Tritiated water (HTO)
  - Uranine

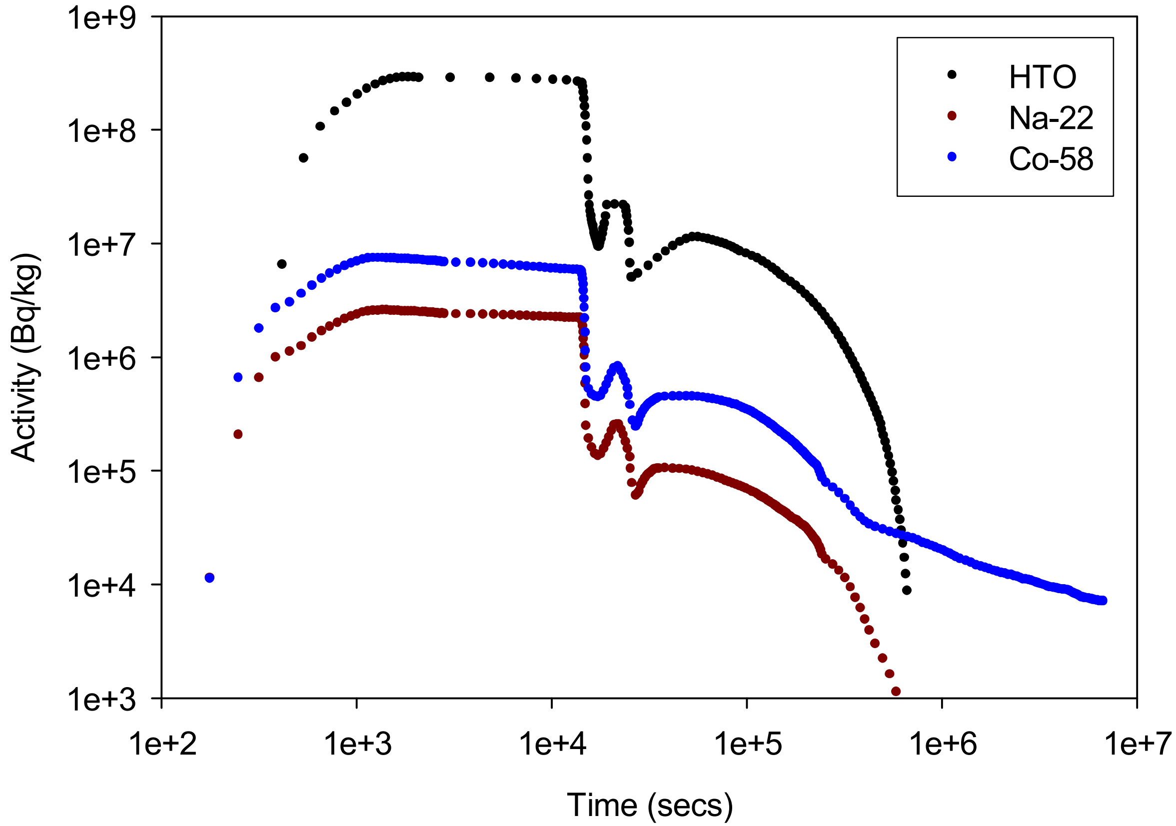
    Non-sorbing
  - Na22
  - Sr85

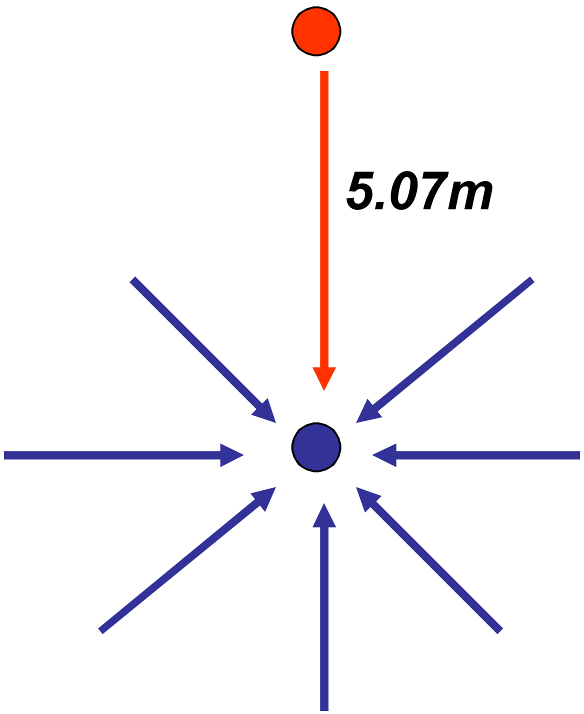
    Weakly to moderately sorbing
  - Rb86
  - Co58

    Strongly sorbing

STT-1b Tracer Tests

Injection histories for three tracers – *Not a square wave*



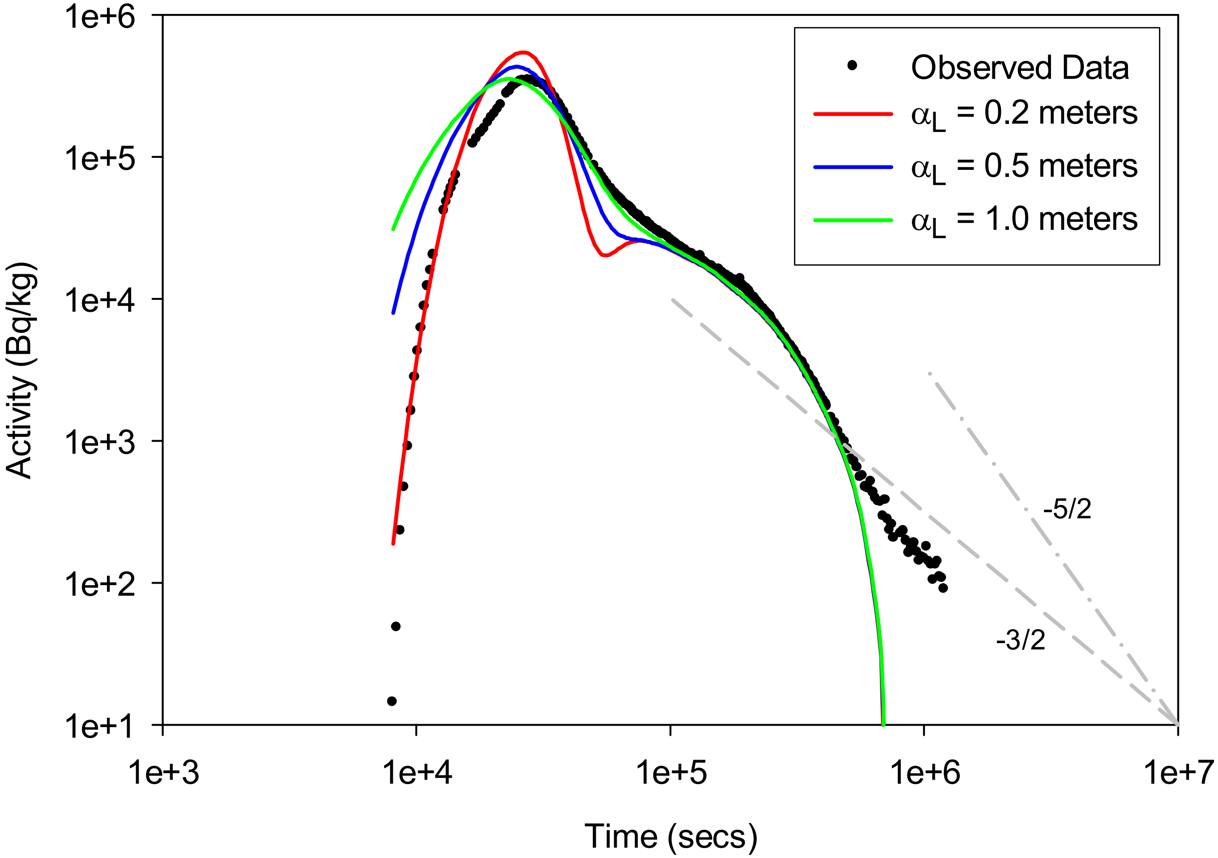


Modeling Approach

- Start with modeling HTO as it is part of the water molecule and is non-sorbing
  - Define all advective parameters from HTO data
- Can breakthrough curves be modeled without mass transfer?
- Can breakthrough curves be modeled with a single rate of mass transfer?
  - Predict other tracers using HTO parameters and laboratory data
- Does consideration of multiple rates of mass transfer improve the models?
- Consistency with geologic conceptual model

Single Porosity Model

Different values of dispersivity (4, 10 and 20 percent of transport distance) change fit to advective portion of breakthrough curve, but don't match late time data



Late-time data have a slope steeper than –3/2 (infinite capacity).

Late-time slope is closer to –5/2

<u>Model fit with:</u>

$\boldsymbol{V_x}$ **= 2.76E-04 m/s**

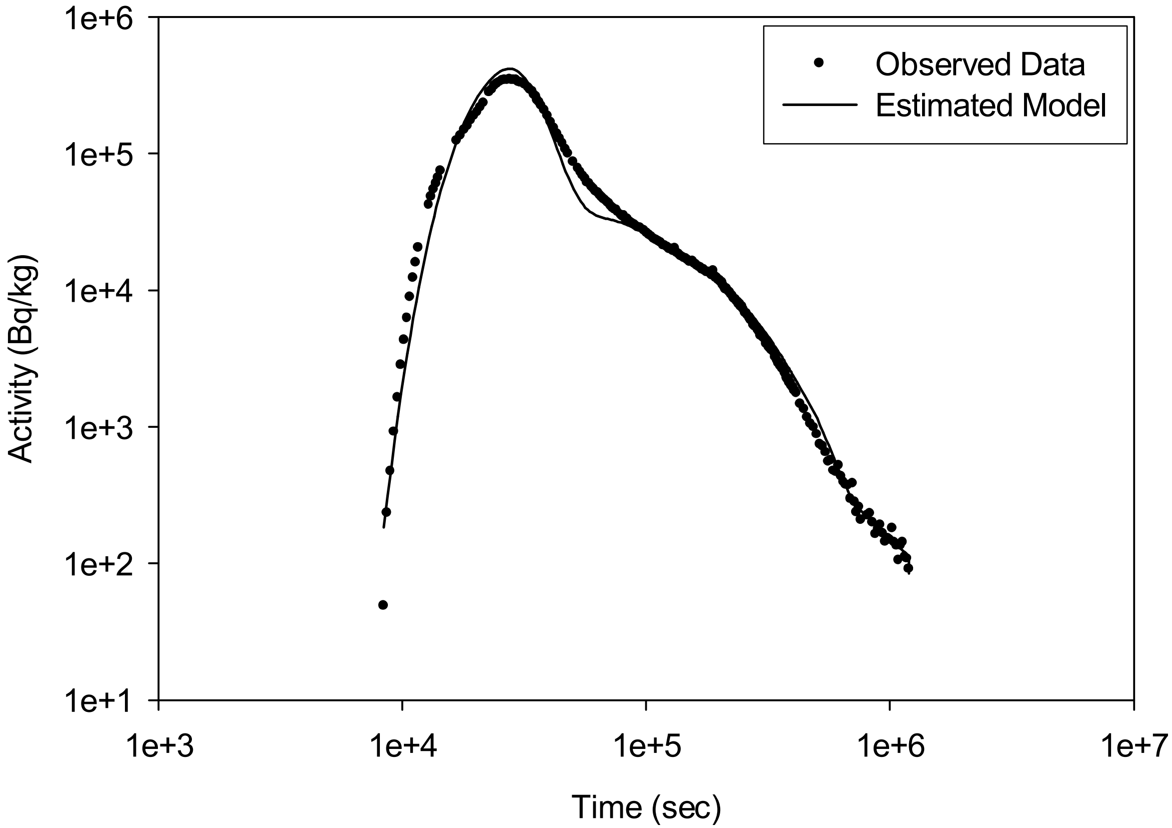
dilute **= 427**

$\boldsymbol{\alpha_L}$ **= 0.2 m**

Single-Rate Mass-Transfer

Adding a single-rate of mass-transfer between the fracture and the matrix allows the model to fit the late time data

Also smoothes out misfits near peak and after peak



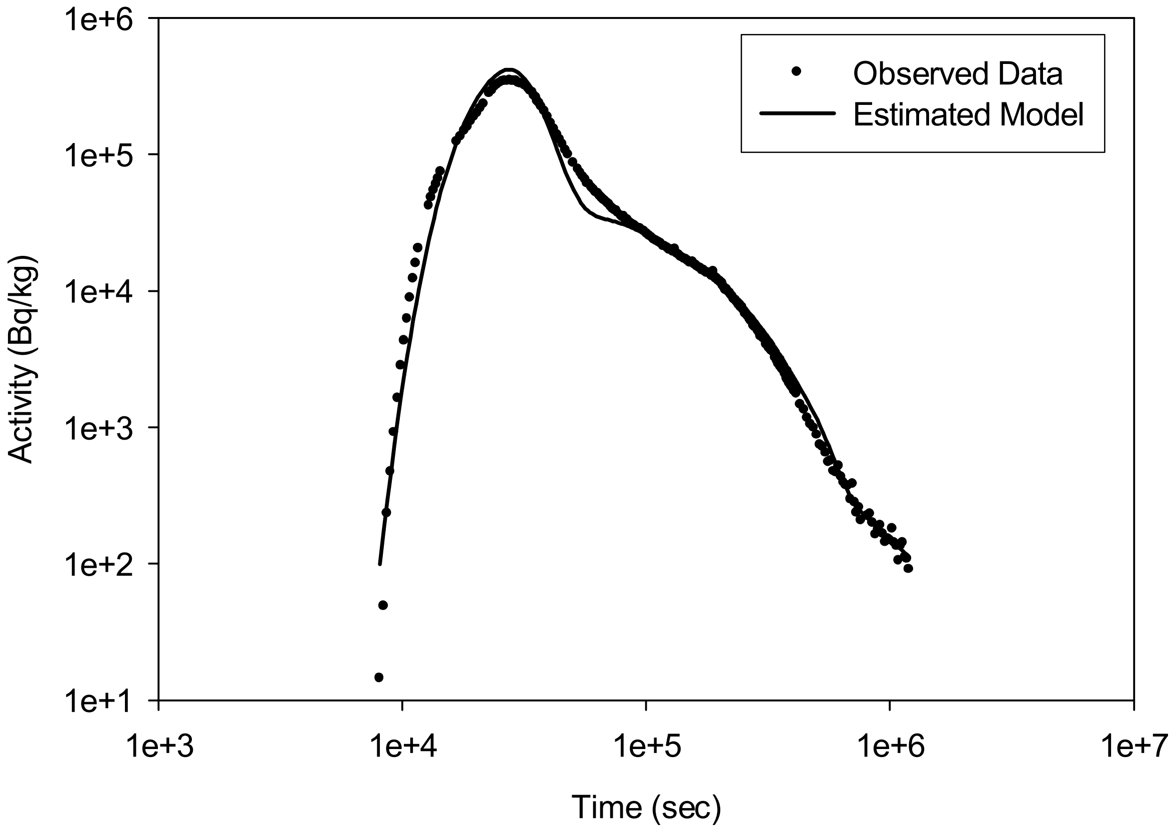
Model fit with:

β_{tot} **= 5.6**

μ **= -16.0 1/s**

Multitrate Mass Transfer

Estimating: V_x, *dilute*, α_L, β_{tot}, μ, σ ***Multi-Porosity Transport***



Model fit with:

β_{tot} = 6.3

μ= -16.3 1/s

σ = 0.4

Comparing Results

Parameters	Single-Rate	Multirate
V_x *(m/s)*	2.76E-04	2.76E-04
dilute	426.7	426.7
α_L *(m)*	0.2	0.2
B_{tot} *(-)*	5.6	6.3
μ *(1/sec)*	-16.0	-16.3
σ *(1/sec)*	0.0	0.4
RMSE	0.26	0.26

Advective-dispersive parameters are held constant for both models

$$RMSE = \sqrt{\frac{1}{N}\sum_{1}^{N}\left(\log 10(C_{obs}) - \log 10(C_{est})\right)^2}$$

Multi-porosity model has additional parameter, but <u>no better fit to data!</u>

Predict Other Tracer Tests

Advective parameters: V_x, *dilute* and α_L are fixed for all tracers

Additionally, β_{tot}, the ratio between porosities is known

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

The diffusion rate coefficient is a function of the ability of the tracer to diffuse into and adsorb to the matrix

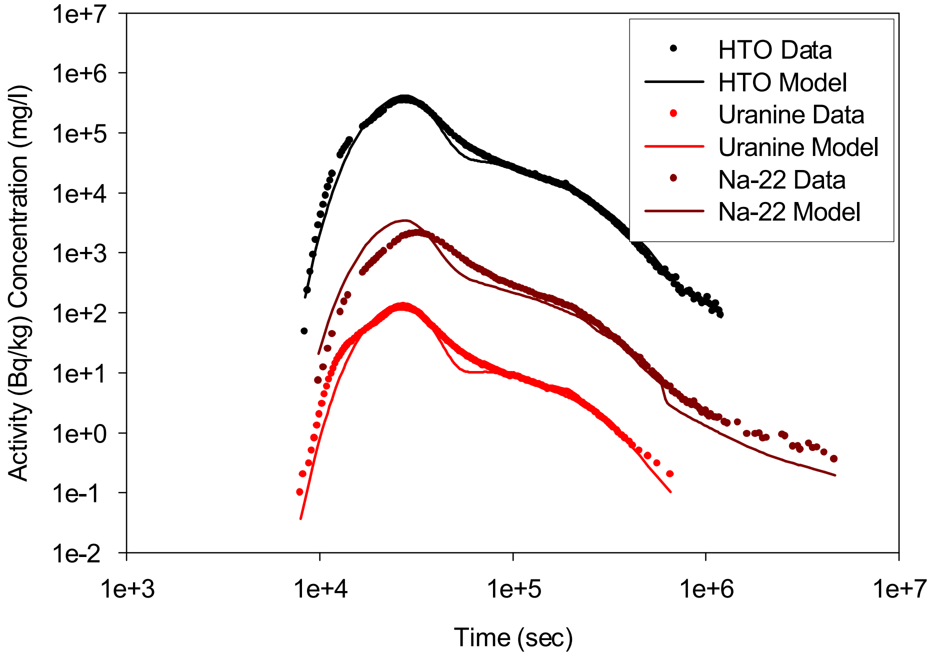
$$\mu = \frac{D_{aq}\tau}{l^2 R_{im}}$$

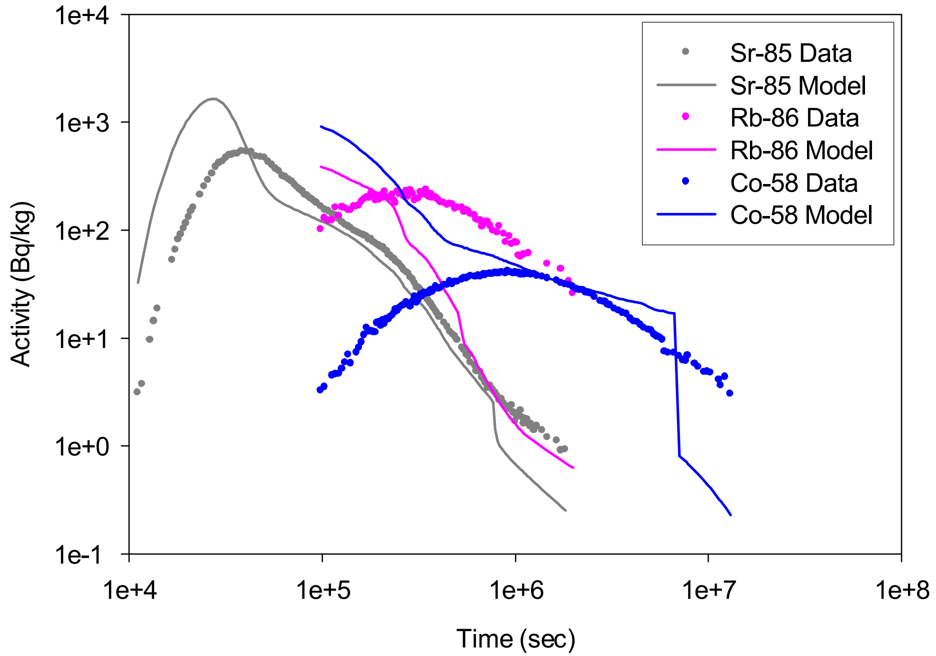
The capacity coefficient is a function of the sorption properties of the fracture wall and matrix

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

Prediction Results

Combining HTO results with laboratory data



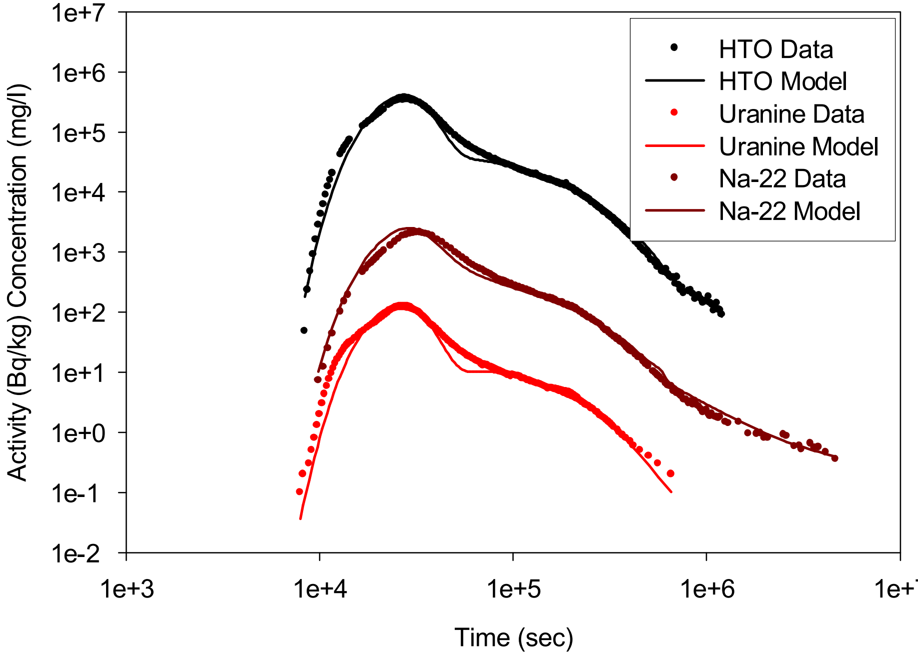


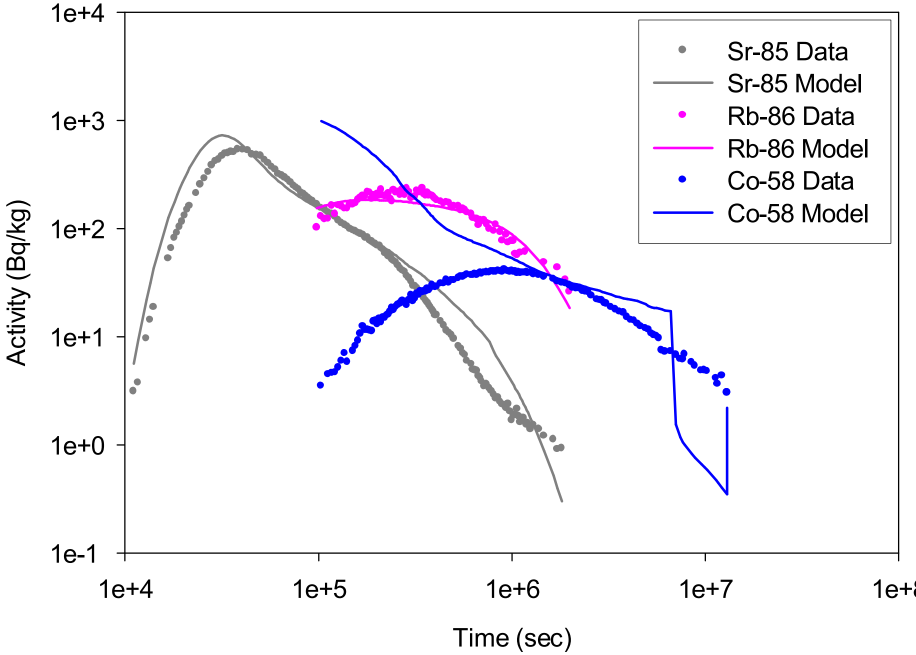
Results are OK for tracers without sorption, but are not accurate for tracers with significant sorption

Wrong parameters (lab data are incorrect)? and/or Wrong processes (Single-rate model is incorrect)?

Single-Rate Estimation Results

Estimating two parameters: mass-transfer rate coefficient, μ, and total capacity, β_{tot}

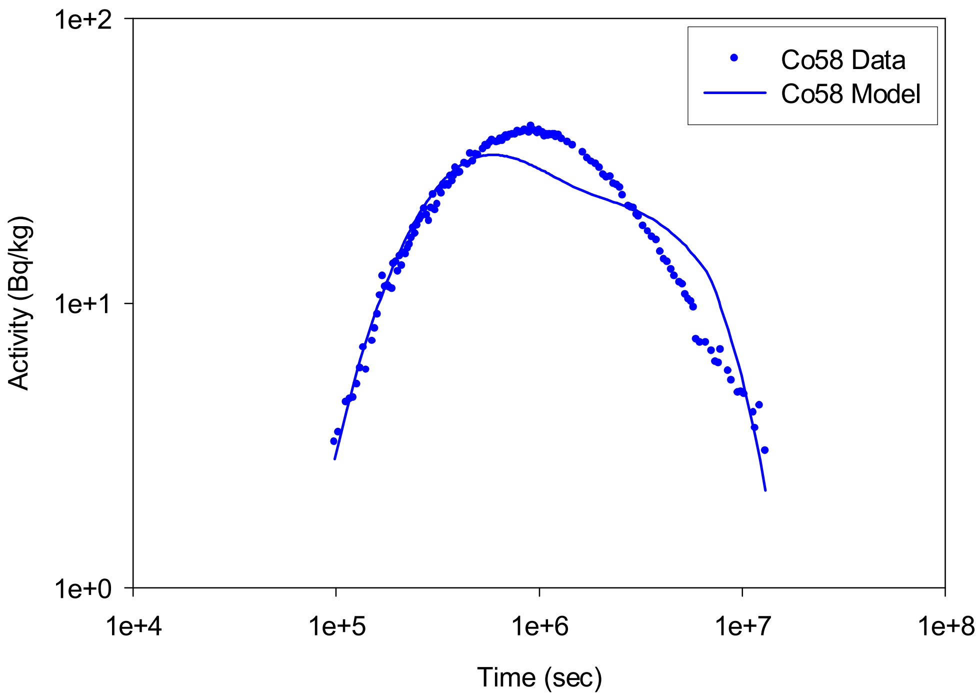




Estimations are considerably closer to observed data than were predictions, especially for more strongly sorbing tracers

Co-58 is still a problem

Revised Co58 Estimation



Two parameter estimations of Co58 results start with activities much higher than observed

Results are indicative of non-equilibrium sorption

Estimate 3 parameters, include *dilute*, to account for less than expected mass recovery

$$dilute = \frac{Q_{pump}}{Q_{inject}} \cdot \frac{1}{RMF}$$

Comparing Single-Rate Models

K_d Values are taken from batch testing in the laboratory for predictions

K_d Values are obtained from estimates of β_{tot} from estimating tracer test breakthrough curves

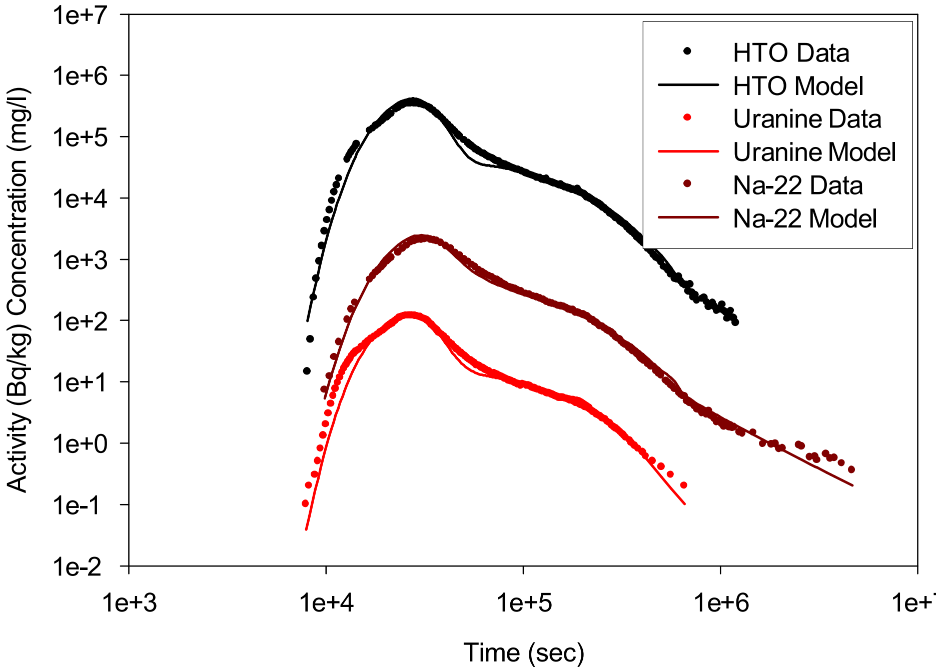
Tracer	Laboratory Kd	Tracer Test Kd
HTO	0.0E+00	0.0E+00
Uranine	0.0E+00	-6.1E-05
Na22	7.0E-06	1.3E-04
Sr85	2.4E-05	1.3E-05
Rb86	3.0E-03	4.4E-04
Co58	1.7E-02	2.6E-03

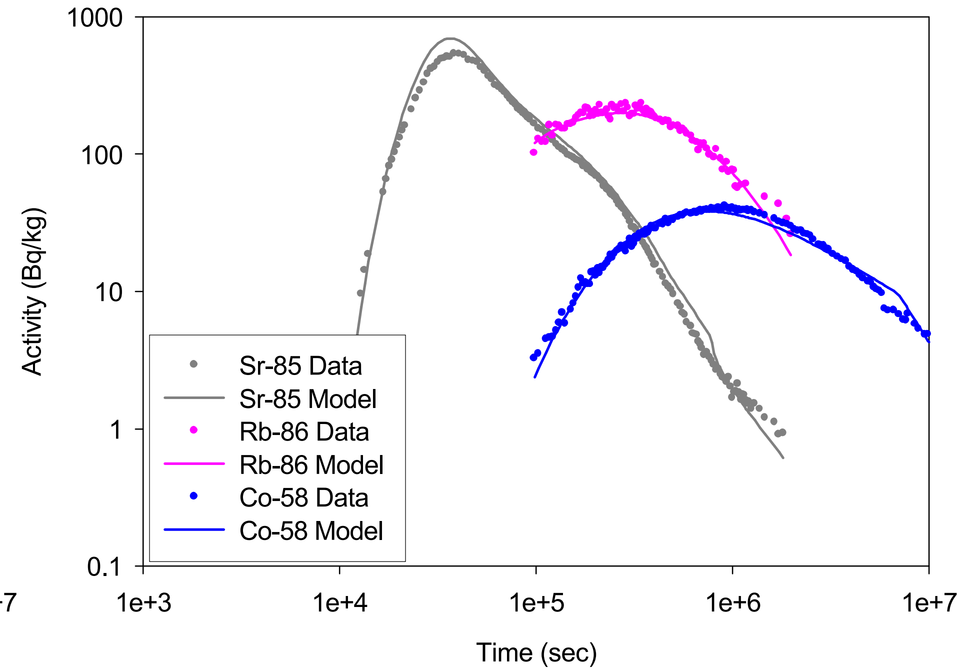
<u>Negative</u> K_d indicates aversion to matrix rock – may be anion exclusion effect

For more strongly sorbing tracers, field estimates are a factor of 2 to 6 lower than lab estimates

Multirate Estimation

Multirate model is used to fit data. μ, σ and β_{tot} are estimated, V_x, *dilute*, and α_L are held constant,





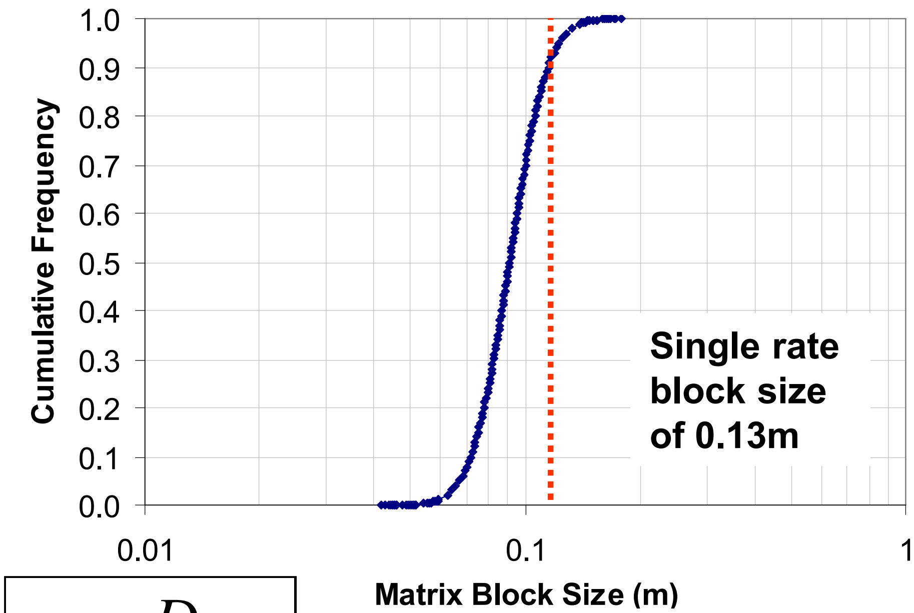
Necessary to also fit *dilute* for Co58 curve

Multirate estimates are better, but are they a significant improvement?

F-test shows all improvements are significant with exception of HTO

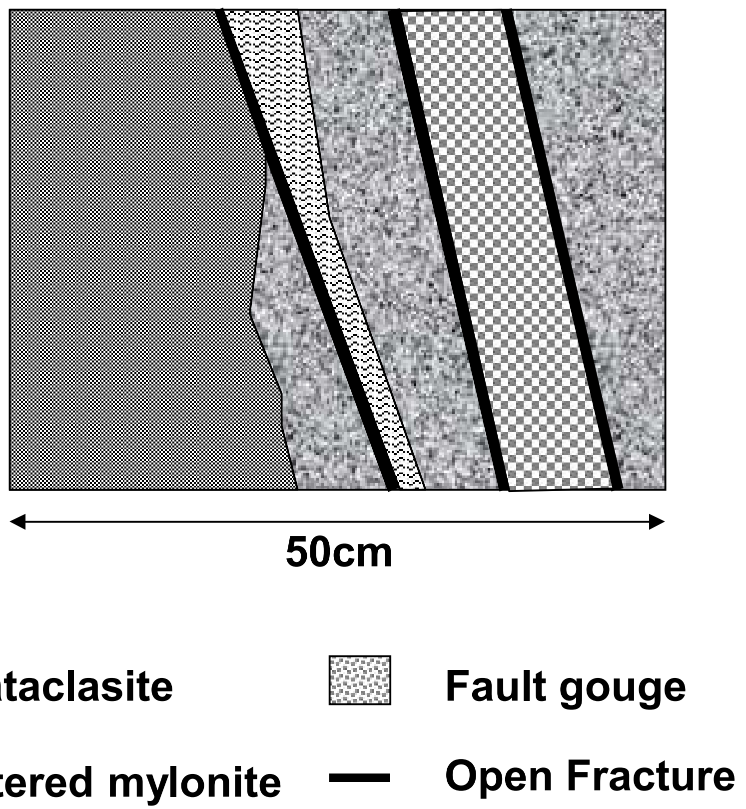
Fit to Conceptual Model

Distribution of mass-transfer rates can be converted to matrix block size distributions

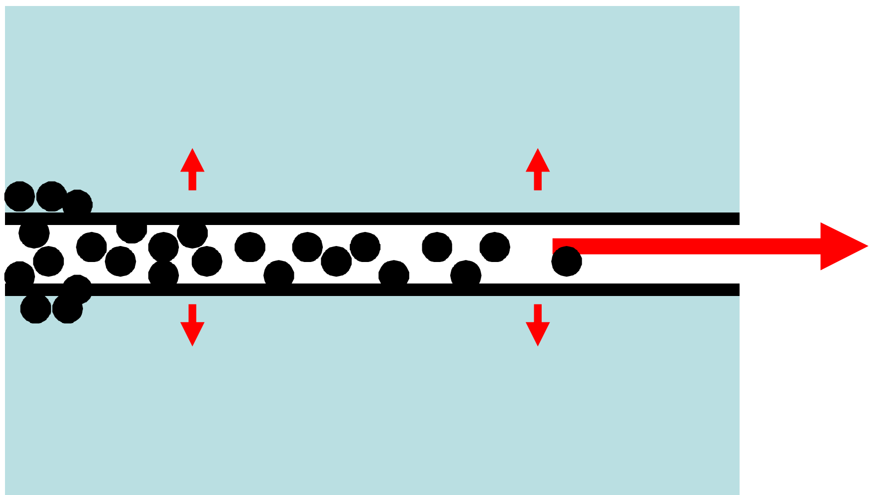


$$\alpha_i = \frac{D_{aq}\tau}{l^2 R_{im}}$$

Block size distribution fits conceptual model of materials within Feature A



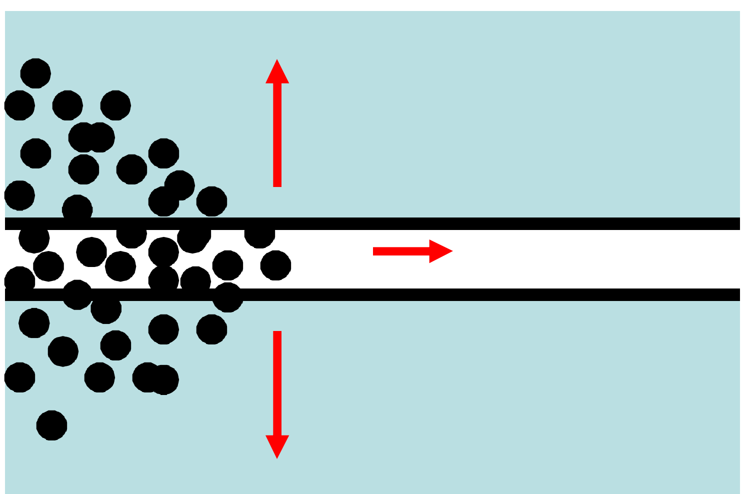
Tracer Testing: Relative Rates



Advection >> Diffusion

Low Damkohler Number

Small fraction of matrix is saturated with solute



Diffusion Rate >> Advection

High Damkohler Number

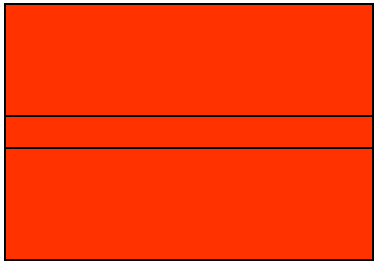
Large Fraction of matrix is saturated with solute

What Can the Test See?

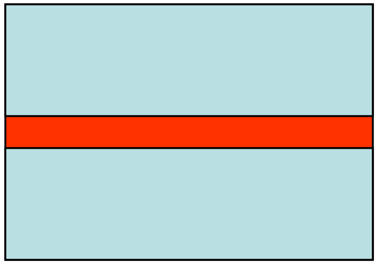
Damkohler number provides ratio of mass-transfer rate to advective rate

$$DaI = 3\alpha_i(1+\beta_i)\frac{LR_m}{v_x}$$

***DaI* >> 1.0 indicates local equilibrium behavior**

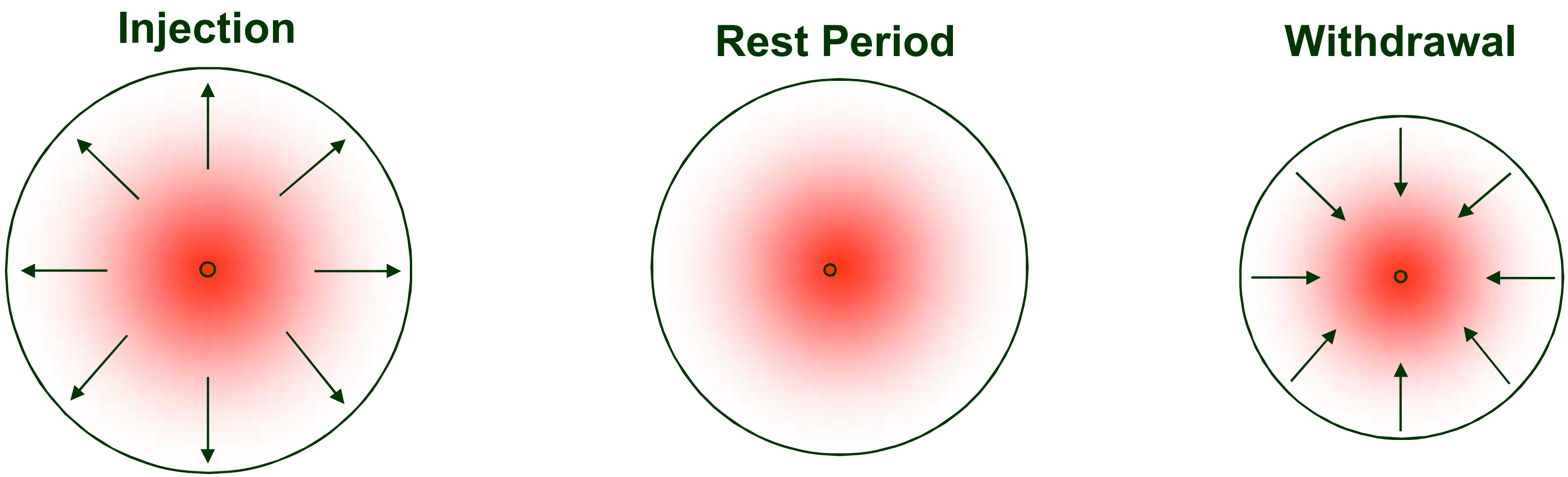


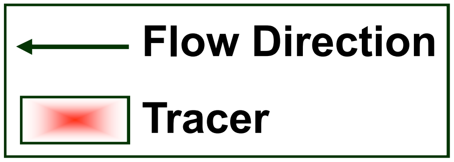
***DaI* << 1.0 indicates single porosity transport**



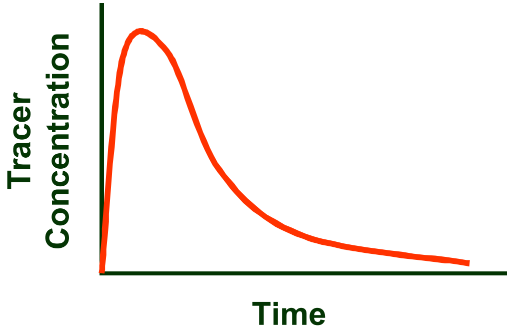
Single-Well Injection-Withdrawal

Single-Well Injection Withdrawal Tests (aka "huff-puff", "push-pull")

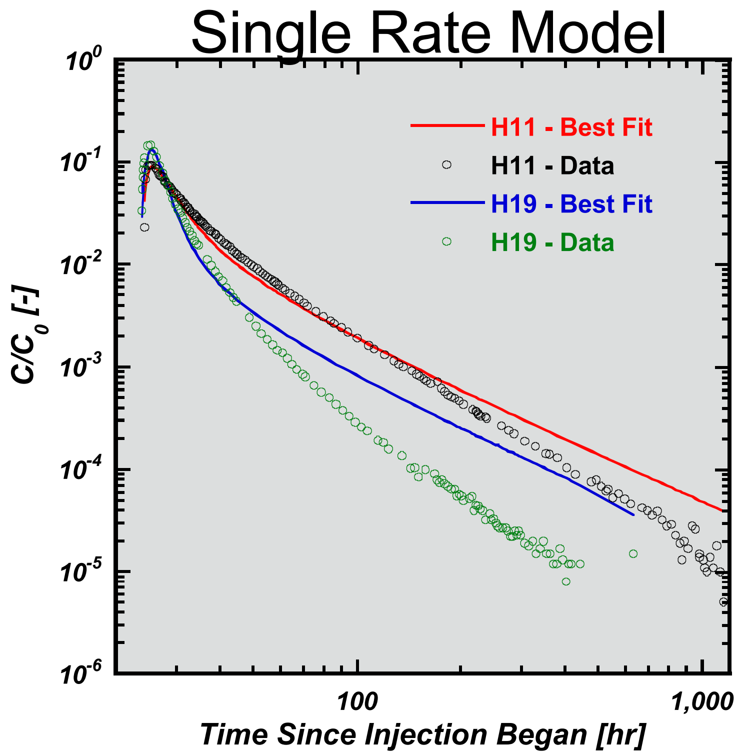




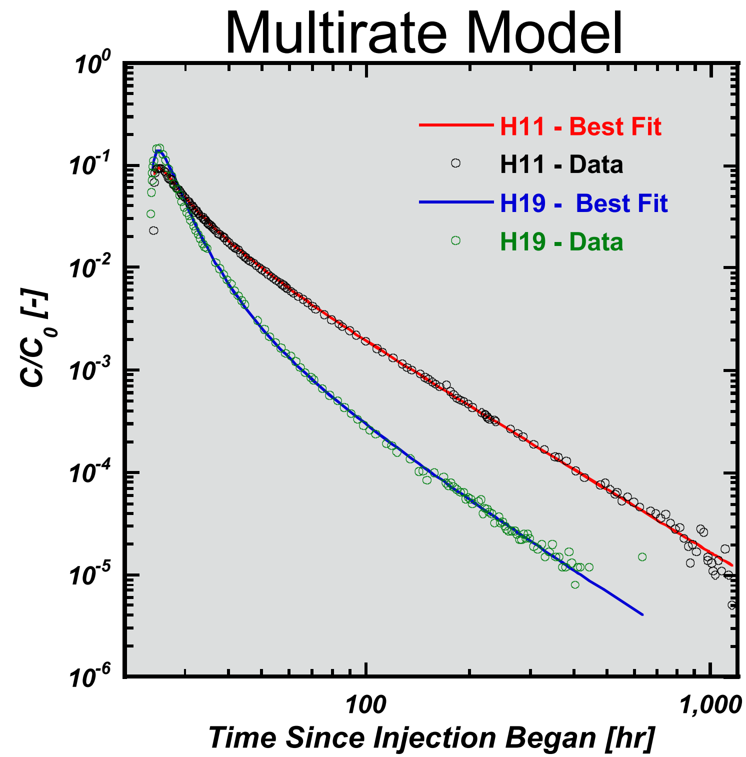
Recovery Curve



WIPP Example: Culebra Tracer Tests

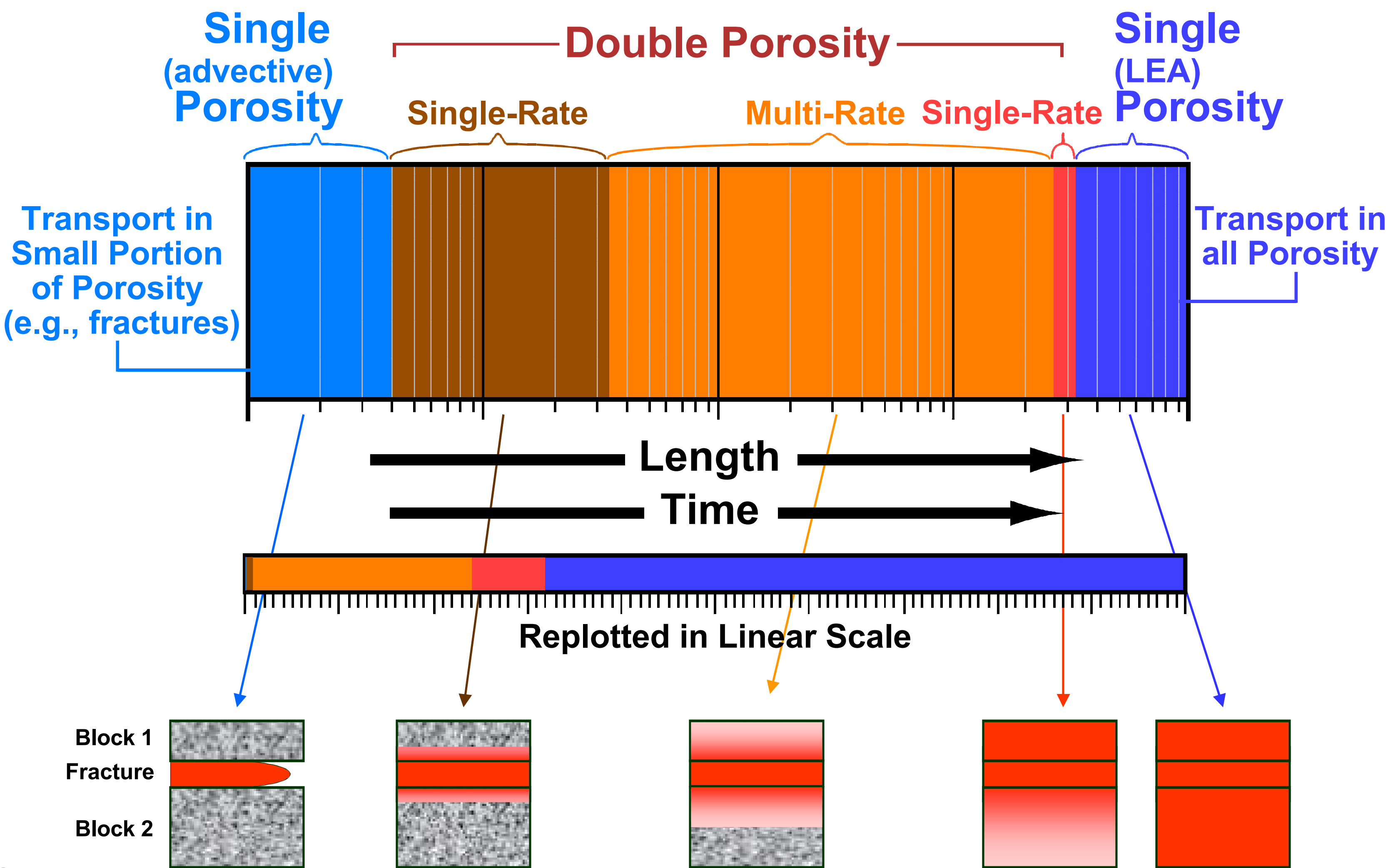


Cannot Capture Tailing Behavior



Provides Excellent Fits Throughout Time Range

Beyond the Tracer Test Scale



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Conclusions

- Signature of mass-transfer processes in tracer test results is unique from dispersive processes
- For Aspo tracer test results:
  - Dispersion alone cannot account for results
  - Single-rate of mass transfer applies to non-sorbing and moderately sorbing tracers
  - Kd values from laboratory are generally too high to predict field results
  - Multiple rate mass transfer model required for strongly-sorbing tracers

Conclusions (Cont.)

- **Aspo tracer test results (Cont.):**
  - **Multirate mass-transfer model provides significantly better fits to tracer data**
  - **Multirate results are consistent with conceptual model of geologic materials within Feature A**

Additional Information

- McKenna, S.A. and J.-O. Selroos, 2004, Constraining Performance Assessment Models with Tracer Test Results: A Comparison of Two Conceptual Models, *Hydrogeology Journal*, 12 (3), pp. 243-256
- Altman, S.J., L.C. Meigs, T. L. Jones and S.A. McKenna, 2002, Controls of Mass-Recovery Rates in Single-Well Injection-Withdrawal Tracer Tests with a Single-Porosity Heterogeneous Conceptualization, *Water Resources Research*, Vol. 38, No. 7,
- Haggerty, R., S. W. Fleming, L.C. Meigs and S.A. McKenna, 2001, Tracer Tests in a Fractured Dolomite. 2. Analysis of Mass Transfer in Single-Well Injection Withdrawal Tests, *Water Resour. Res.*, 37 (5), pp. 1129-1142.
- McKenna, S.A. and L.C. Meigs, and R. Haggerty, 2001, Tracer Tests in a Fractured Dolomite 3. Double Porosity, Multiple-Rate Mass-Transfer Processes in Two-Well Convergent Flow Tests, *Water Resour. Res.,* 37(5), pp. 1143-1154.
- Haggerty, R., S. A. McKenna, and L. C. Meigs, 2000, On the late-time behavior of tracer test breakthrough curves, *Water Resour. Res.,* 36(12), pp. 3467-3479.

BACKUP SLIDES

Quantify Small-Scale Diffusivity

- Rock samples with fault gouge, altered and unaltered diortie from Kamaishi Mine in northern Japan
- What is the variability in diffusivity values in a small sample?
- Is there a relationship between porosity and diffusivity?

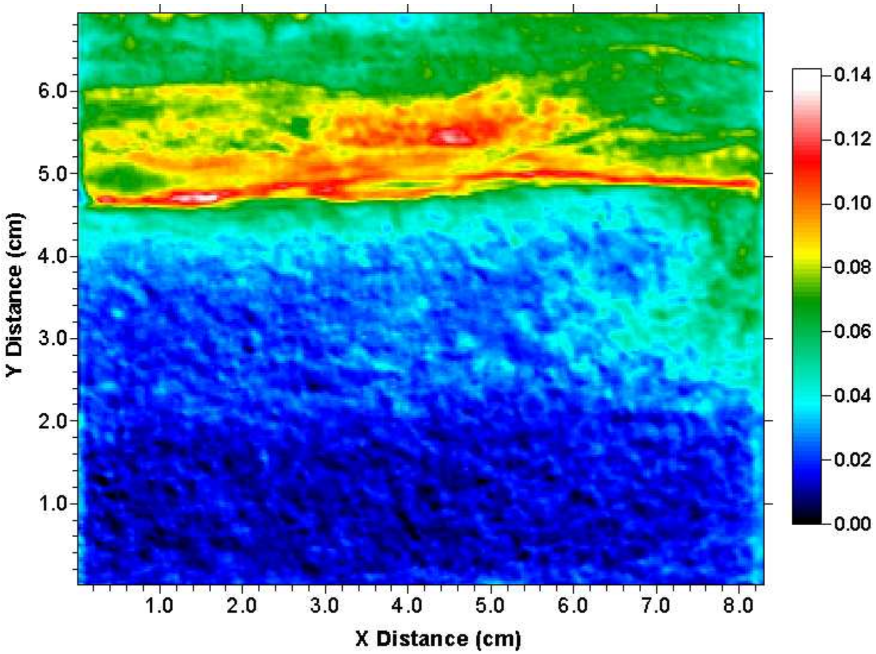
Imaging Diffusion

- X-ray adsorption imaging technique developed at Sandia:
  - *Tidwell and Glass, 1997; Water Resour. Research*
- Approach
  - Sample is placed between X-ray source and film
  - Iodide tracer adsorbs X-rays (linear relation between adsorption and iodide concentration)
  - X-ray film is developed and digitized
  - Difference between dry and saturated images gives porosity

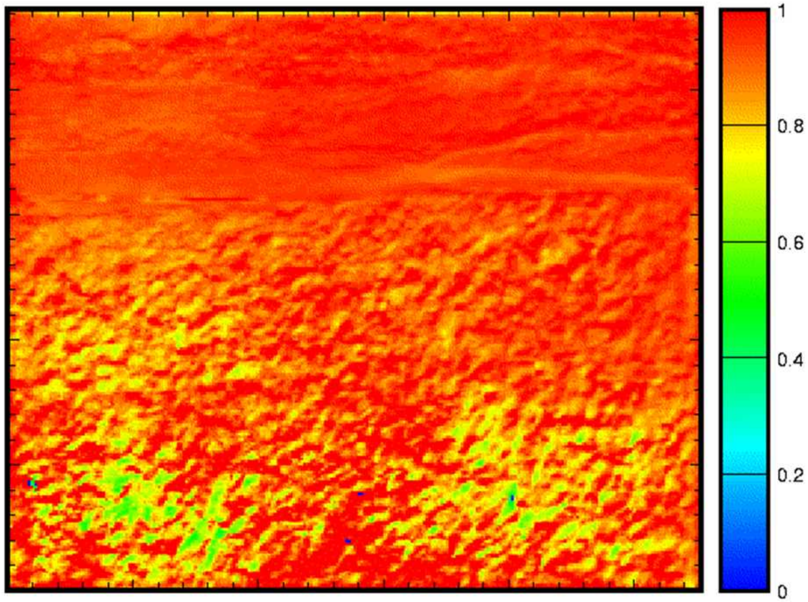
Laboratory Evidence

X-ray Transmission imaging of KI tracer in fracture filling material from granites at the Kamaishi Mine in Japan

Porosity



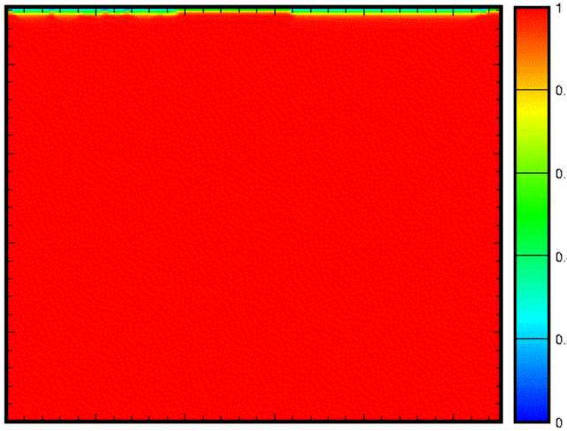
C/Co



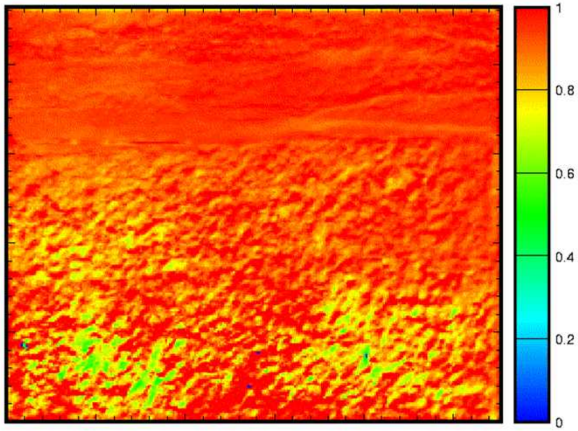
Images out to 10 days with log spacing of times

KC1c-FF Results Animations

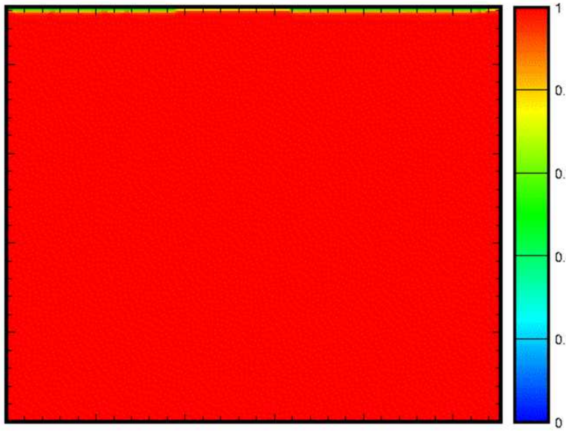
Optimized Porosity Zonation



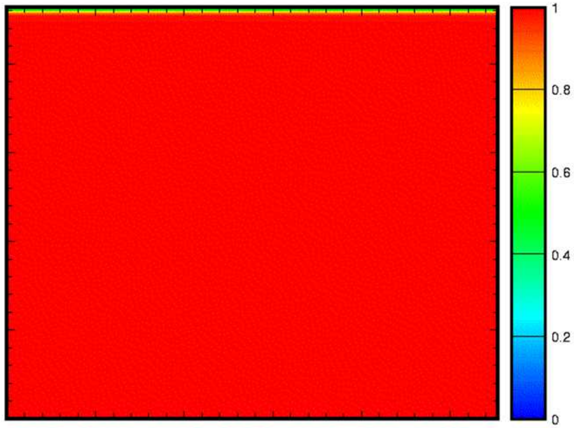
Experimental Data



Initial Porosity Zonation

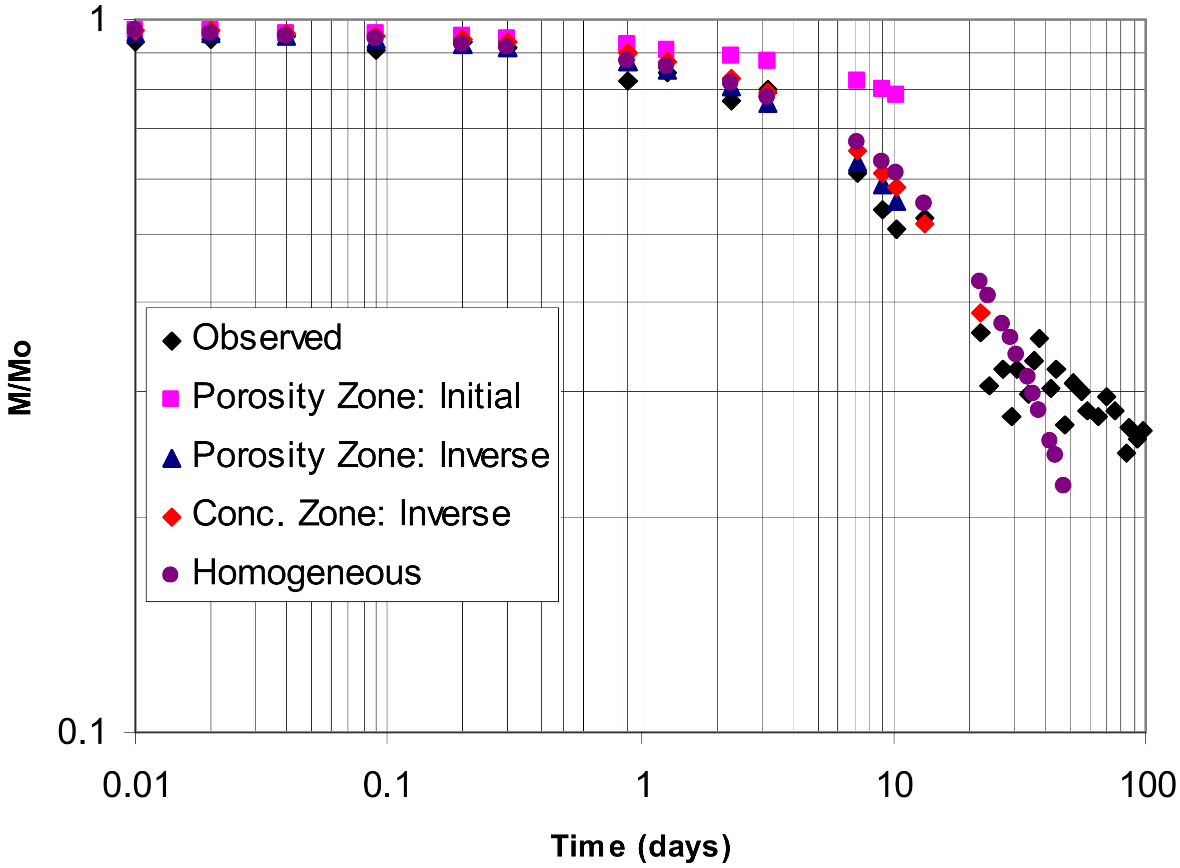


Concentration Zonation



Best fit to observed data is provided by the concentration zonation

KC1c FF Results



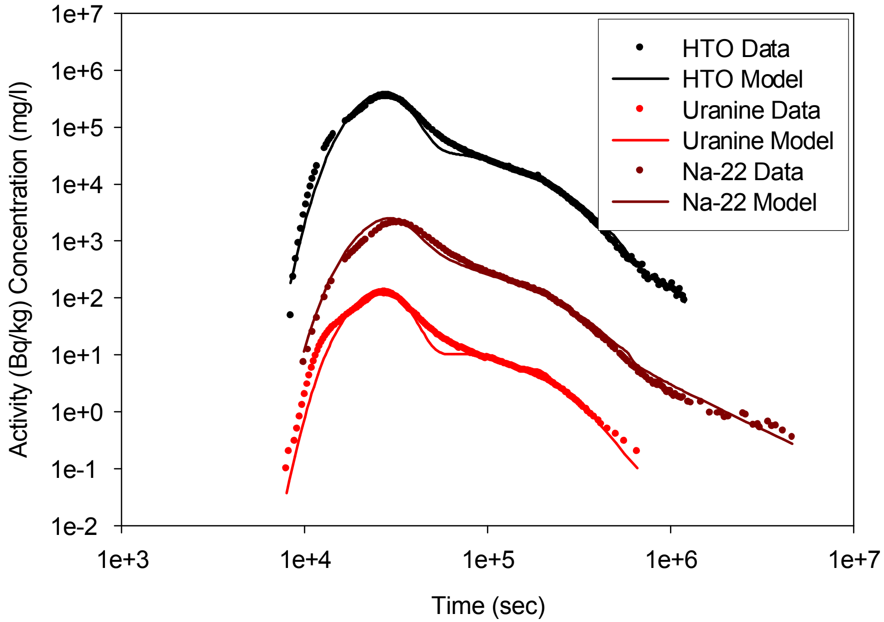
All optimized solutions improve on initial estimates

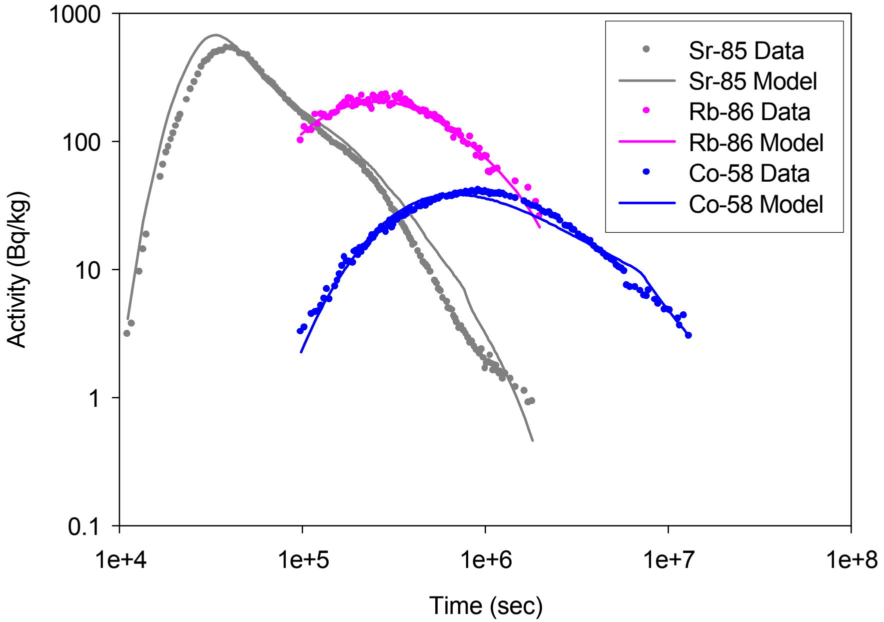
Change in slope of data is noisy, could possible be real due to very low De in low porosity material

Double Porosity Fits

Model fits with single mass-transfer rate and no surface sorption

SPHERICAL FITS

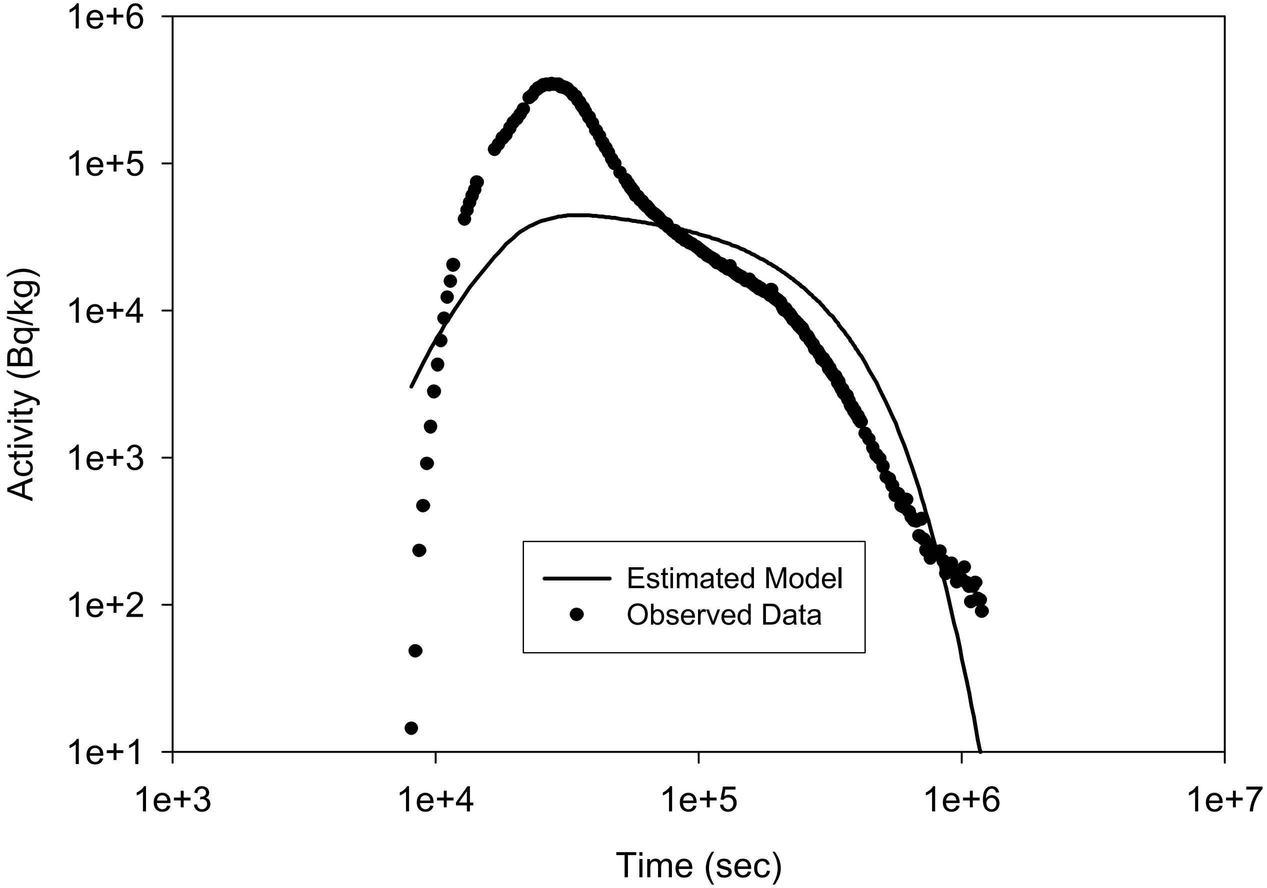




Model has difficulty with Uranine, Sr-85 and Co-58

Single-Porosity Model

Estimating: ***V_x*, *dilute*, *α_L*** ***Single Porosity Transport***

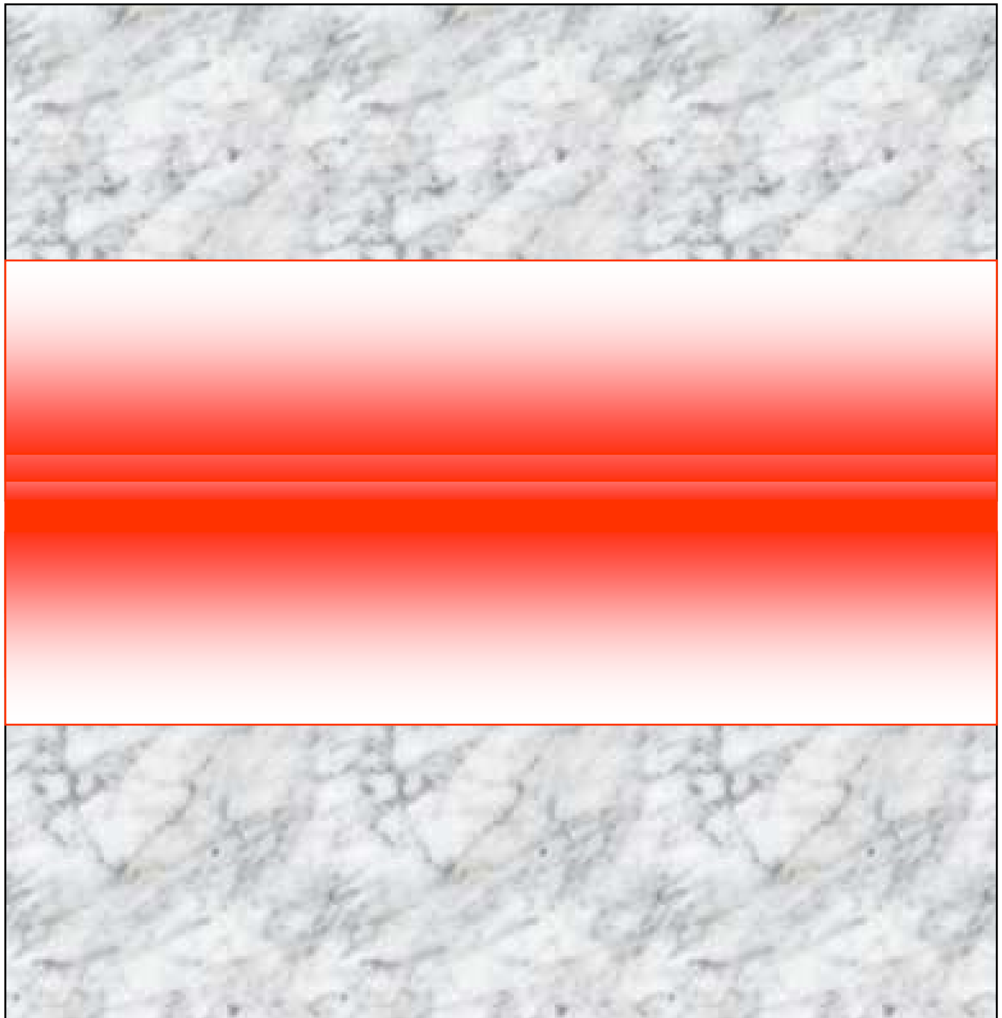


Comparing Results

Parameters	Single Porosity	Multi-Porosity
V_x (m/s)	6.55E-05	2.76E-04
dilute	644.7	426.7
α_L (m)	6.0	0.2
B_{tot} (-)	NA	6.30
μ (1/sec)	NA	-16.3
σ (1/sec)	NA	0.37
RMSE	1.15	0.26

$$RMSE = \sqrt{\frac{1}{N}\sum_{1}^{N}\left(C_{obs} - C_{est}\right)^2}$$

Matrix Block Size



Diffusion into an infinite medium means that the concentration front does not encounter any boundary conditions that limit the rate of diffusion

The fracture surface area to matrix volume ratio is small

Matrix Block Size

Diffusion into a finite medium means that the concentration front does encounter boundary conditions that limit the rate of diffusion

The fracture surface area to matrix volume ratio is relatively larger

Culebra Dolomite

Porosity Types:
Fractures
Vugs
Intercrystalline
Interparticle