

# Designing Tracer Tests to Assist in Formulating Conceptual Site Models, Site Characterization, and Estimation of Chemical Transport Properties

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USEPA-USGS Fractured Rock Workshop

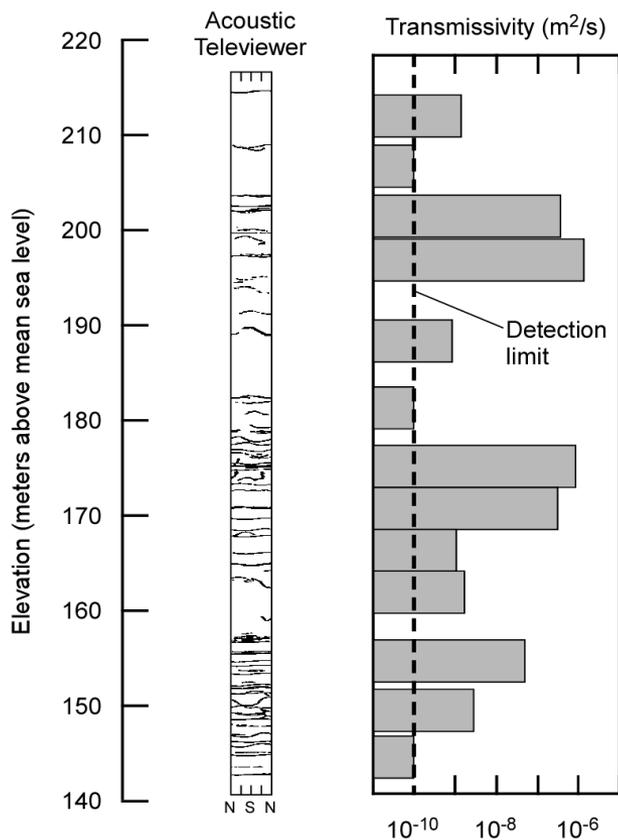
EPA Region 10

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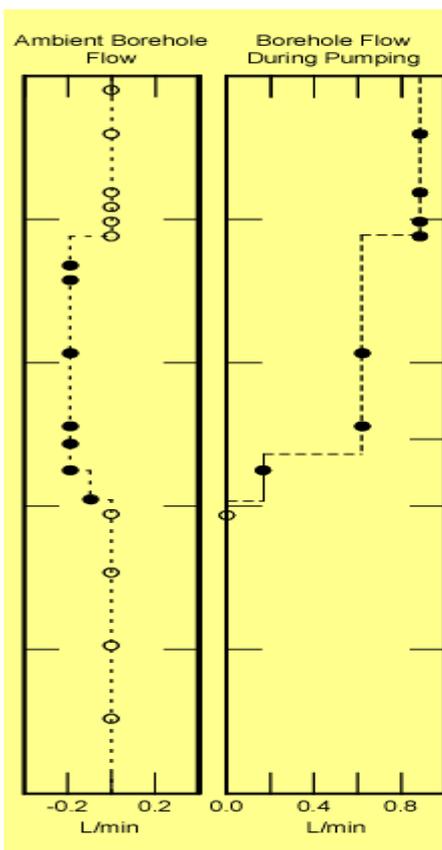


# Permeable groundwater flow paths are identified by single-hole and cross-hole hydraulic tests, borehole geophysical logging methods, etc.

Single-hole hydraulic tests conducted in borehole H1



Borehole flowmeter survey conducted in borehole H1

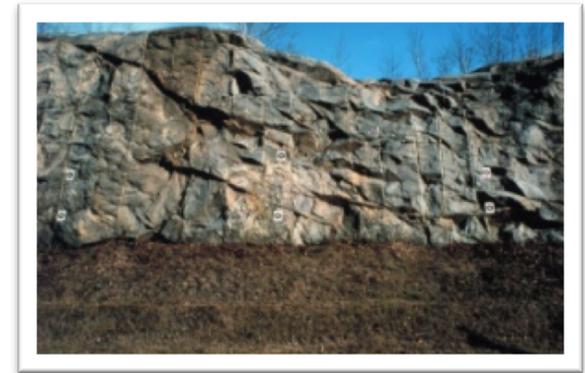
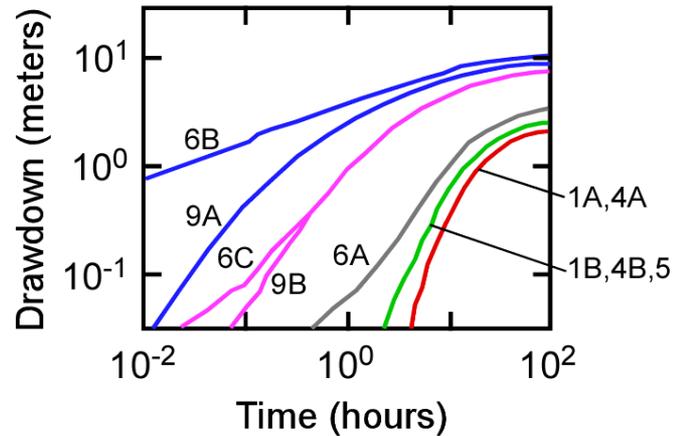
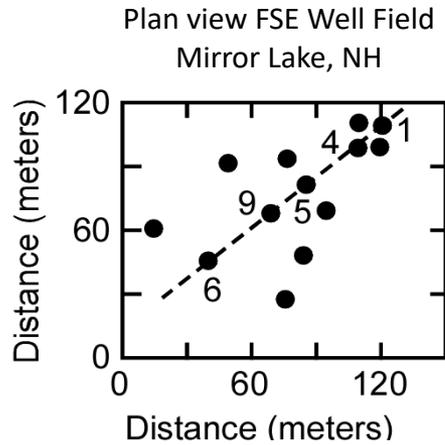


Straddle packers isolate a section of the borehole to conduct single-hole hydraulic test

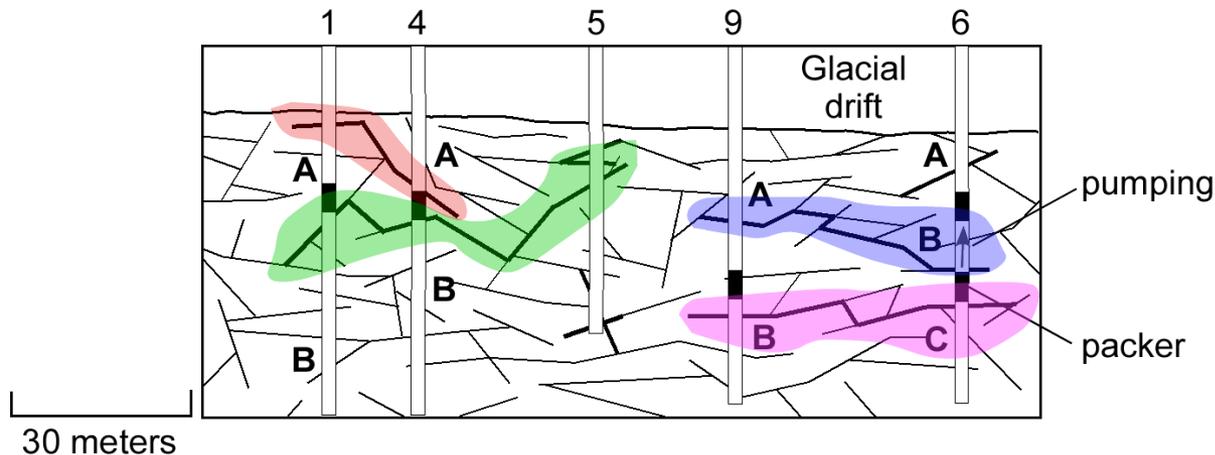
Granite and schist along road cut near Mirror Lake, NH



# Permeable groundwater flow paths identified by cross-hole hydraulic tests



Road cut near Mirror Lake, NH  
Granite and schist



# Why . . . tracer testing . . . at sites of groundwater contamination. . .

Permeable groundwater flow paths define pathways for contaminant transport and (volumetric) groundwater fluxes

Important information about the magnitude of processes affecting fate and transport of contaminants is missing from the characterization of groundwater pathways and fluxes

For example. . . relating the volumetric (Darcy) flux to the groundwater velocity. . .

$$q = -K \frac{dh}{dx}$$

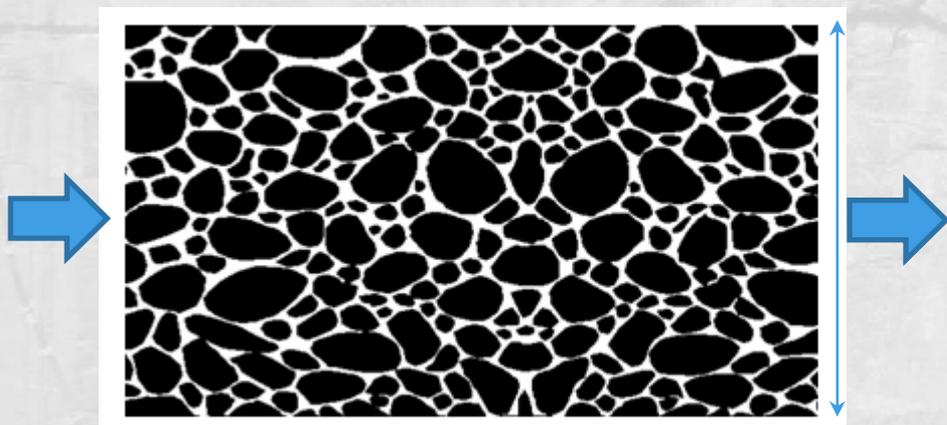
Darcy flux – volumetric flux per over entire x-sectional area

$$v = -\frac{K}{n} \frac{dh}{dx}$$

Groundwater velocity – advective movement of a constituent – only through area occupied by fluid

$K$  – hydraulic conductivity

$n$  – porosity – void volume per total volume



# Why . . . tracer testing . . .

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## What's important ?

- Confirming groundwater flow paths identified from hydraulic tests
  - Quantify chemical residence time and dilution
  - Chemical processes attenuating contaminant concentrations and residence time (e.g., diffusion, sorption/desorption)
  - Processes affecting contaminant transformations (abiotic and biotic processes)
  - Processes affecting particulate, colloidal, or pathogen migration
  - Estimate residual DNAPL in subsurface
- 

## Why ?

- Conceptual models of contaminant retention and release
- Evaluating contaminant longevity
- Designing amendment injections, and treatments of source zones and plumes

# What is a tracer test ?

*Basic premise: Introducing a chemical constituent (particulate, etc.) into the groundwater flow regime and monitoring its spatial distribution or arrival to infer processes that affect the fate and transport of the tracer in the subsurface. . .to infer behavior about contaminants of interest. . .*

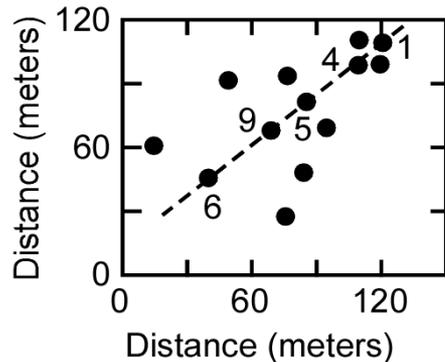
*Operation: (1) **Observations and interpretations of contaminants** in the groundwater flow regime can be used as “tracer” tests. . .provided that (time-varying) groundwater flow regime can be reconstructed. . .(time- and spatially-varying) contaminant introduction into the subsurface can be reconstructed*

*(2) **Controlled “tracer” tests** conducted under ambient groundwater flow conditions or under hydraulic perturbations. . .a known quantity of a “tracer” is introduced into the groundwater flow regime*

*(3) **Observations and interpretations of “environmental” tracers** introduced into the groundwater through atmospheric deposition (e.g.,  $^3\text{H}$ , He,  $\text{SF}_6$ , chlorofluorocarbons, etc.). . .may not be able to discern “site” scale fate and transport processes. . . most likely appropriate for regional flow regimes*

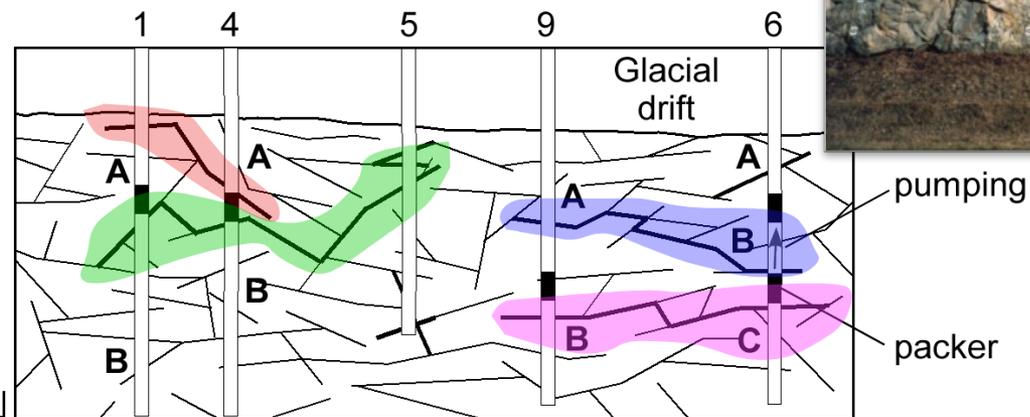
# What is a tracer test ? (continued)

*Interpretation: Knowledge of the groundwater flow regime is a critical component of quantitative interpretations. We recognize the complexity of groundwater flow in fractured rock. . . simplified interpretations of site-scale groundwater flow regime (e.g., linear or radial flow) may lead to erroneous interpretations. . . however, we are unlikely to know intricacy of fracture connections. . . but, significant hydraulic features should be incorporated in interpretation of the tracer test. . .*



Plan view FSE Well Field  
Mirror Lake, NH

30 meters



*Relevance: Are the hydraulic and chemical conditions of the tracer test similar to the conditions of interest?*

## Some Considerations in the design of tracer tests in fractured rock aquifers

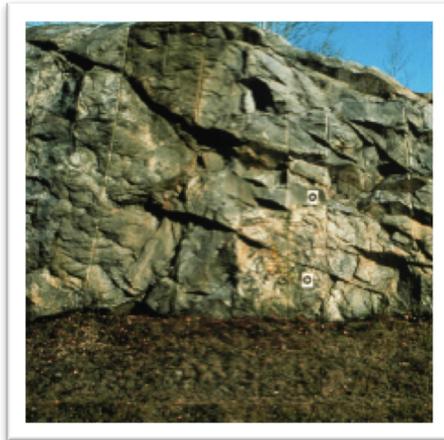
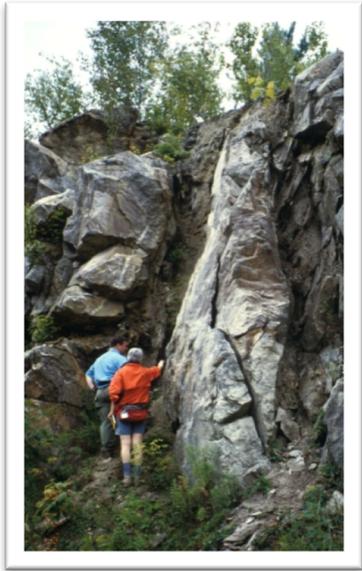
- Tracer tests can be designed over hours, days, months or years, depending on groundwater velocity, monitoring locations, attenuation processes, etc.
- Many types of tracers are available to quantify processes of interest (inert, reactive, varying free-water diffusion coefficients, dissolved gases, bacteria, colloids, microspheres, etc.)
- Maintaining the geochemical signature of the ambient groundwater (oxic, anoxic, fluid density)
- Effect of fluid density in structured media
- How much tracer mass must be added to register and interpreted a response at monitoring locations over the duration of the test? Preliminary estimates of dilution and attenuation are difficult to derive in fractured rock *(In many cases, tracer tests are not run; they are re-run!)*
- Designing injection apparatus at land surface, and apparatus in injection and monitoring boreholes—maintaining geochemical conditions of ambient groundwater; volume of fluid in boreholes may be large relative to fracture volume; borehole volume may dilute tracer responses

# Recall: Fractured rock characterized by hierarchy of void space

*Granite and schist, Mirror Lake Watershed  
Grafton County, New Hampshire*



Iron-hydroxide precipitate staining the rock matrix (primary/intrinsic rock porosity)



Fractures exposed on a road cut (fracture porosity)

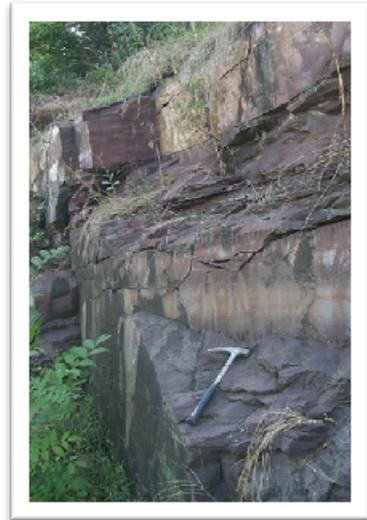
Fault zone exposed on a road cut

Identify the key features and processes affecting contaminant migration in the “hierarchy of void space”

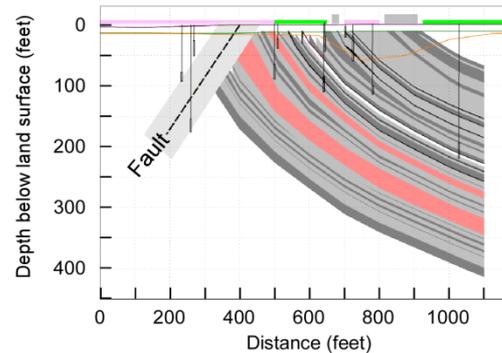


Residual wetting of rock core (primary/intrinsic rock porosity)

*Lockatong Mudstone,  
West Trenton, New Jersey*

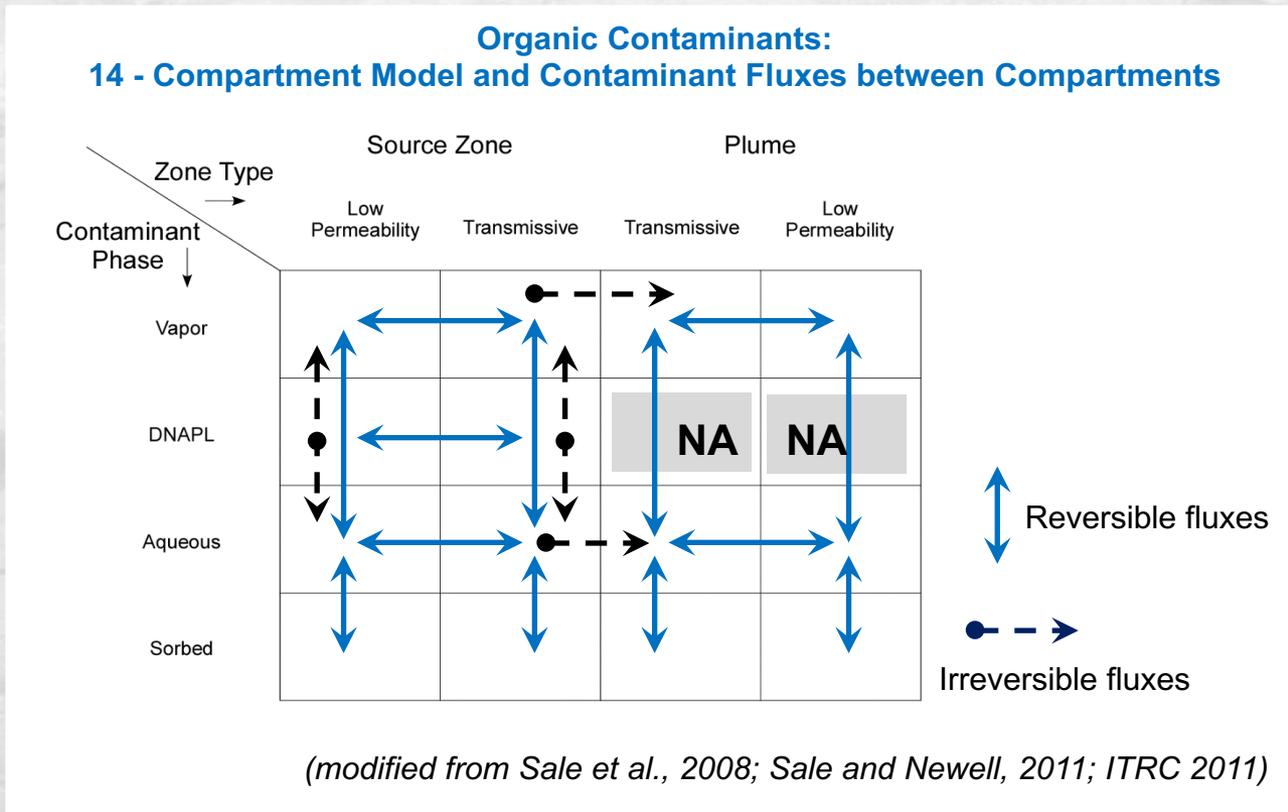


Fractures parallel and perpendicular to bedding (fracture porosity)



Schematic cross section perpendicular to bedding showing fault zone location

It helps to “compartmentalize” our thinking about Conceptual Site Models. . .



- Conceptualize processes that affect contaminant “storage” and contaminant fluxes
- What “reservoirs” are being interrogated by the tracer test? Over what physical dimension and over what time scale?

# What is a quantitatively successful tracer test?

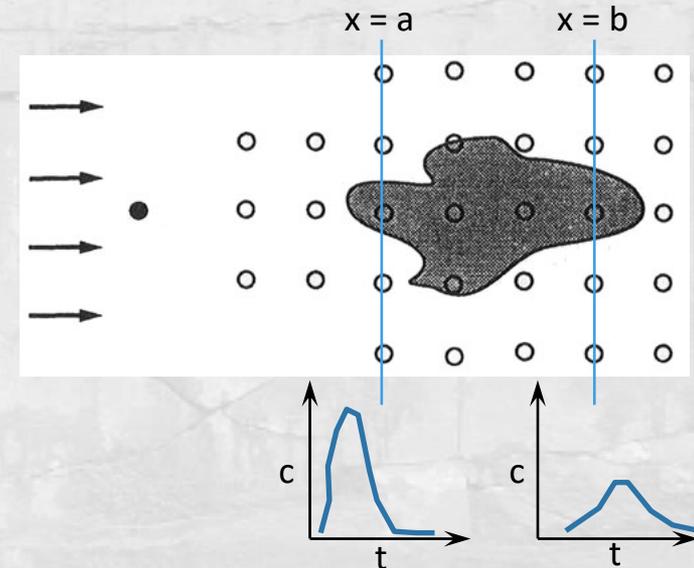
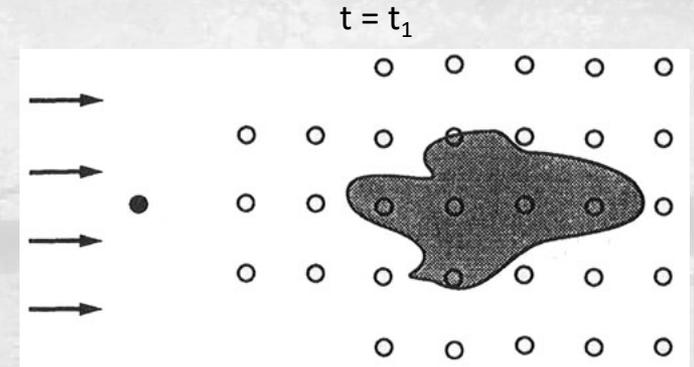
. . .where the groundwater flow regime is known (or assumed), interpretation of the tracer response leads to estimation of those transport properties governing the fate and transport of the tracer. . .

## Spatial distribution

- At  $t = 0$ , a known tracer mass injected
- At  $t = t_1, t_2, \dots, t_n$ , identify the spatial distribution of mass in the formation
- Accounting for all of the injected mass at each snapshot in time ( $t_1, t_2, \dots, t_n$ )

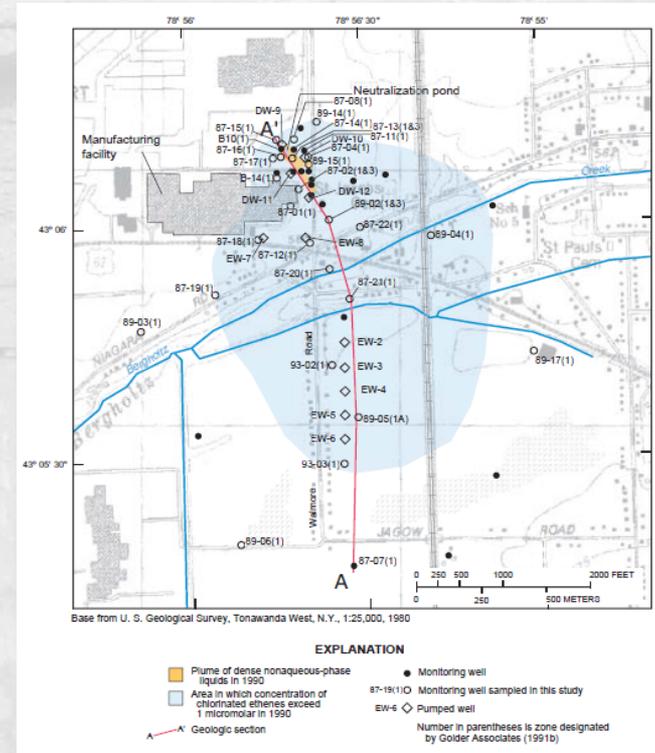
## Mass arrival

- At  $t = 0$ , a known mass injected
- At a boundary,  $x = a, x = b$ , identify mass crossing the boundary for  $t > 0$  (breakthrough curve)
- Accounting for all the injected mass crossing the boundary (breakthrough curve at  $x = a, x = b$ ) ?



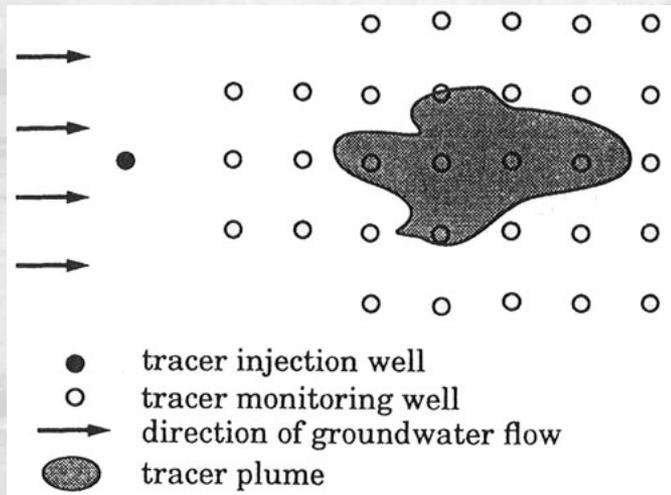
# Observations and interpretations of “contaminants”

- Interpretations of contaminant plumes over 100’s of meters (plume characteristics) – usually, monitoring wells interpreted as if interrogating a single groundwater flow path
- Estimate (1) groundwater velocity, (2) attenuation processes (diffusion between mobile and immobile groundwater, biological attenuation). . . e.g., Twin Cities Army Ammunition Plant (MN) [sandstone], Bell Aerospace Textron Wheatfield Plant (NY) [dolomite]
- Processes can be quantified using observations at successive times (e.g., degradation)
- Difficult to differentiate between attenuation processes (e.g., matrix diffusion, microbial degradation). . . may need other “tools” for this purpose (e.g., isotopic analyses)
- Difficult to infer processes over 10’s of meters (e.g., source zone) where groundwater flow paths are convoluted and monitoring locations are not sufficient to characterize processes along groundwater flow paths



# Controlled tracer tests – ambient flow regimes

Natural gradient tests – tracers injected, migrating by ambient groundwater



NRC 1996



LeBlanc 1991

MA Military Reservation, Cape Cod, MA  
Glacial outwash, unconsolidated sand and gravel

Successfully conducted in unconsolidated porous media. . . installation of monitoring wells is inexpensive. . .

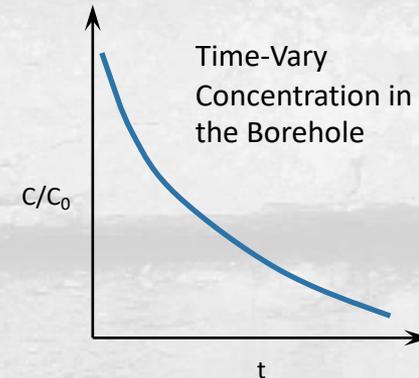
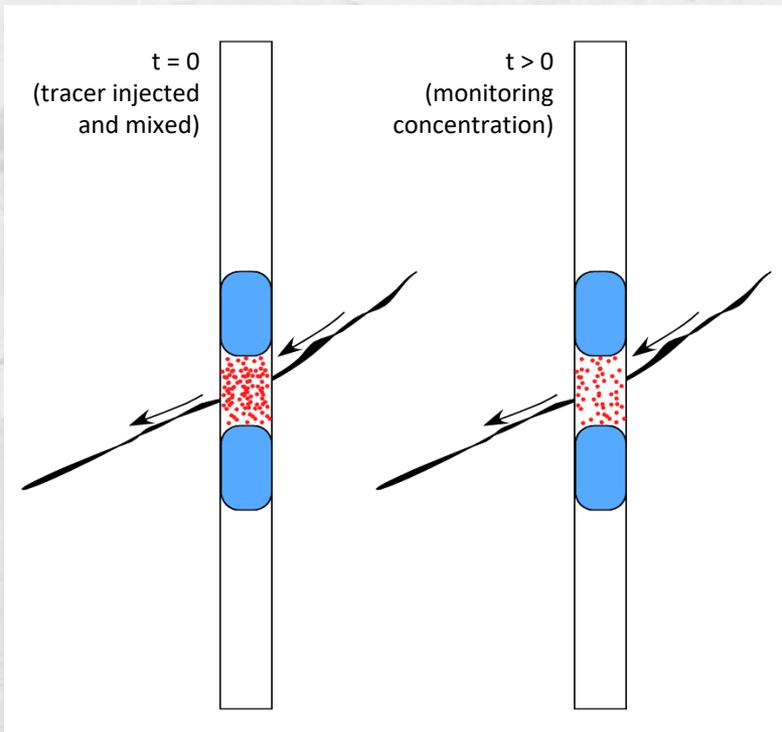
Sparse monitoring locations and convoluted groundwater flow paths in fractured rock – unlikely to lead to a quantitatively successful test in groundwater flow regimes that do not have focused groundwater discharges

Qualitative results from natural gradient tests identify “connections”. . . e.g., dye tracing in karst aquifers between sinkholes and springs. . .

# Controlled tracer tests – ambient flow regimes

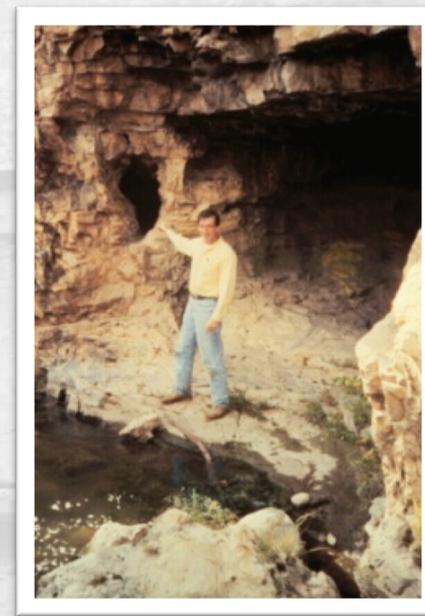
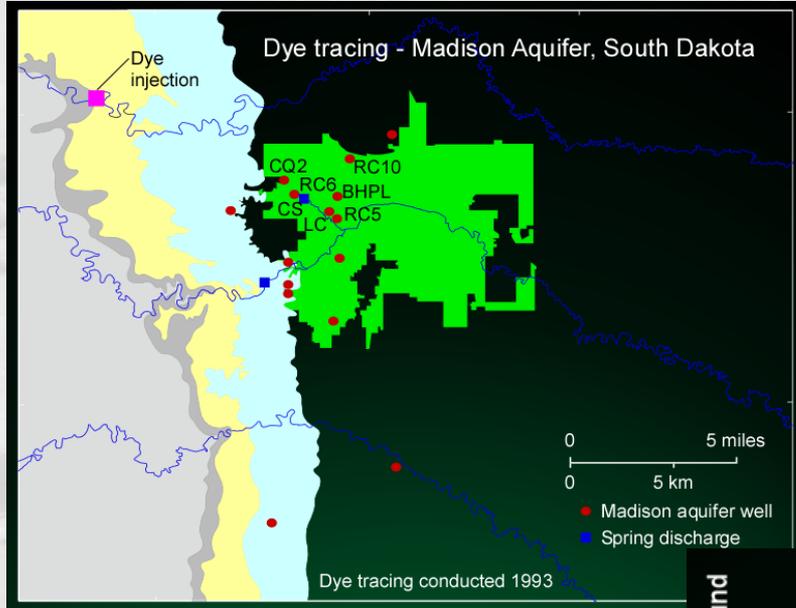
Natural gradient tests – tracers injected, migrating by ambient groundwater

## Single-hole, Point Dilution Test

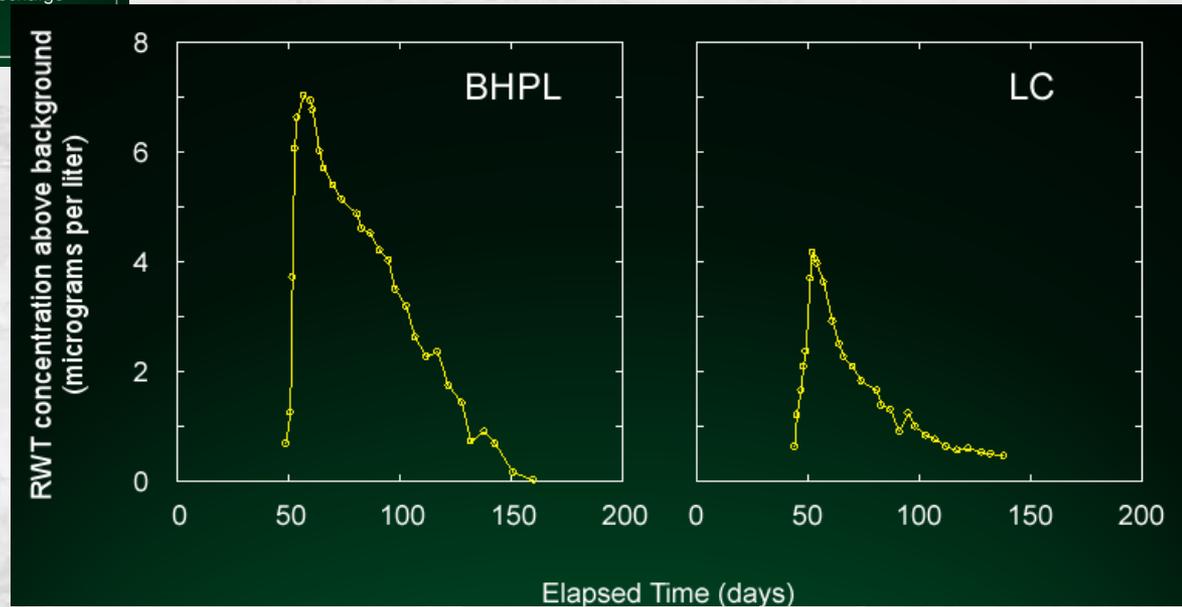


- Usually conducted with an ionic tracer. . . *in situ* monitoring using specific conductance probes. . .
- Local groundwater flux responsible for dilution may not be representative of advective groundwater conditions at other locations. . .
- Ambiguities in interpretation. . . first developed for application in unconsolidated porous media. . . what is the x-sectional area in borehole attributed to discrete fractures intersecting boreholes?

# Controlled tracer tests – ambient flow regimes



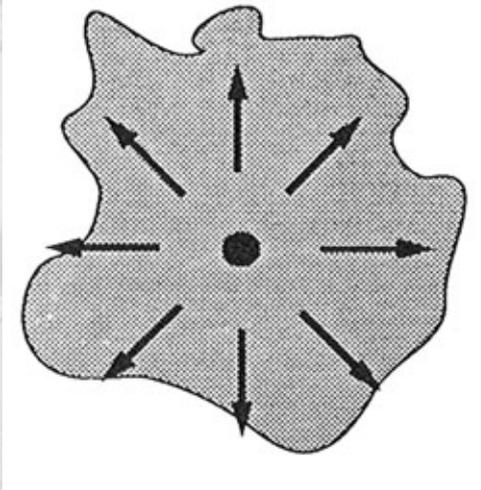
Tracer arrival at selected production wells (> 10 km from tracer injection) in the Madison Limestone



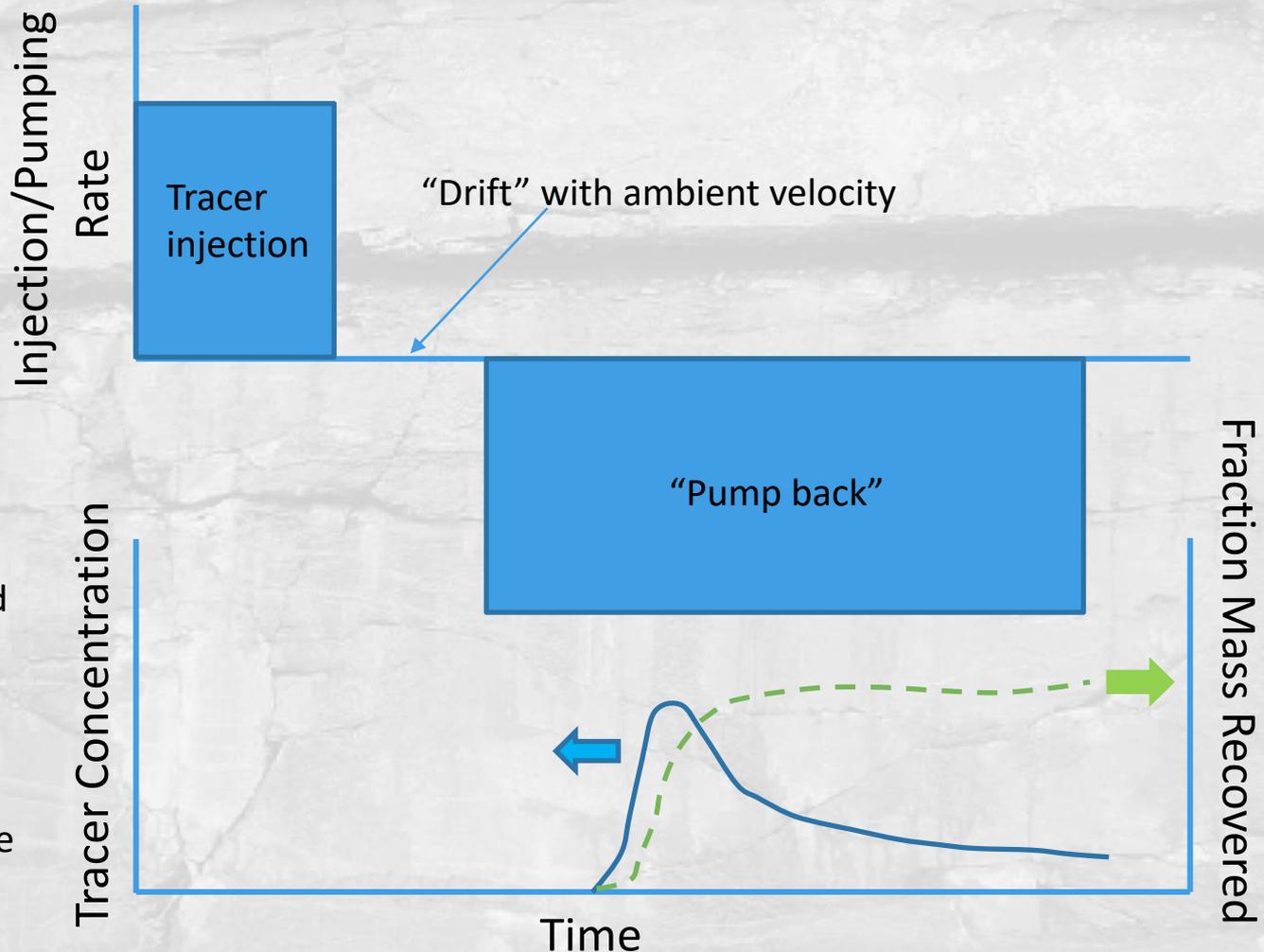
# Controlled tracer tests – Hydraulically stressed conditions

. . .pumping from one or more locations to establish a groundwater flow regime where tracer can be recovered following it's injection. . .

Single-hole tracer tests:

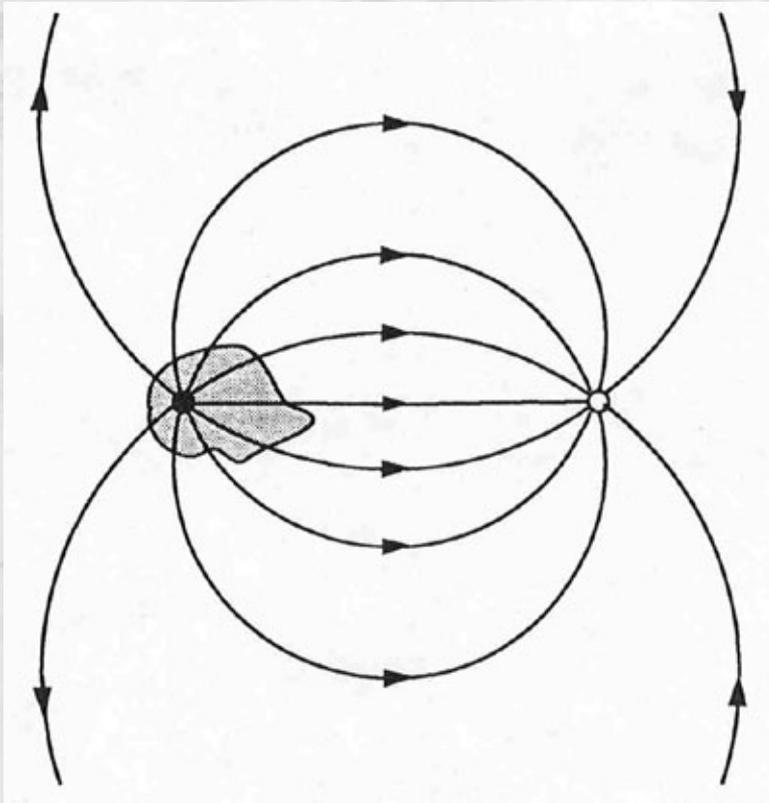


- Conducted in a single fracture or closely spaced fractures intersecting borehole
- Transport processes and properties (single fracture and rock matrix) local to borehole



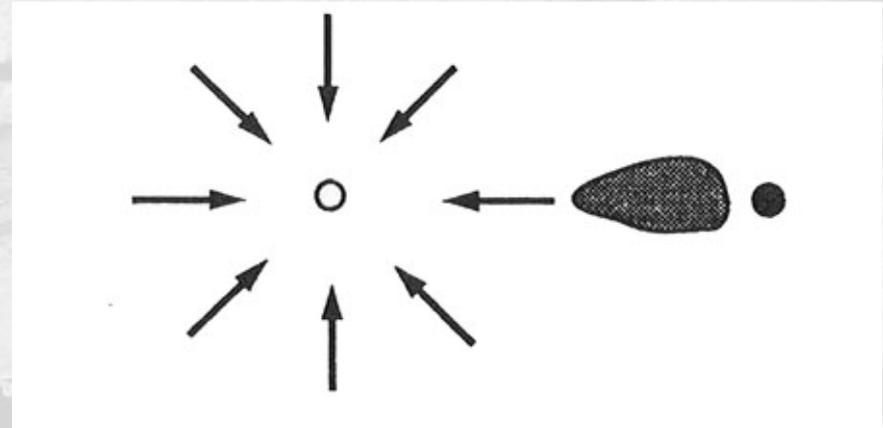
# Controlled tracer tests – Hydraulically stressed conditions

Cross-hole or multiple-hole tracer tests:



Doublet test: Continuous pumping and injection locations

- Conducted with or without recirculation
- Conducted with or without pulse injection of tracer
- Conducted with different pumping and injection rates (e.g., weak dipole)

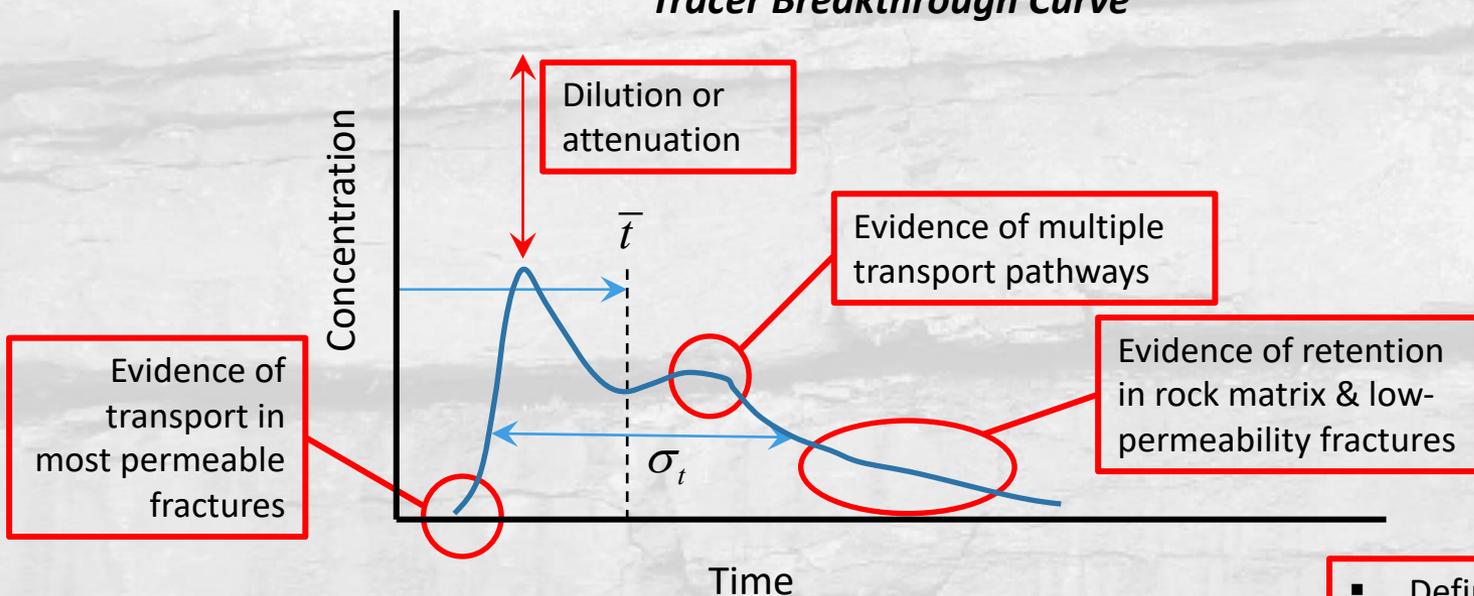


Converging test: Continuous pumping and (finite) pulse injection

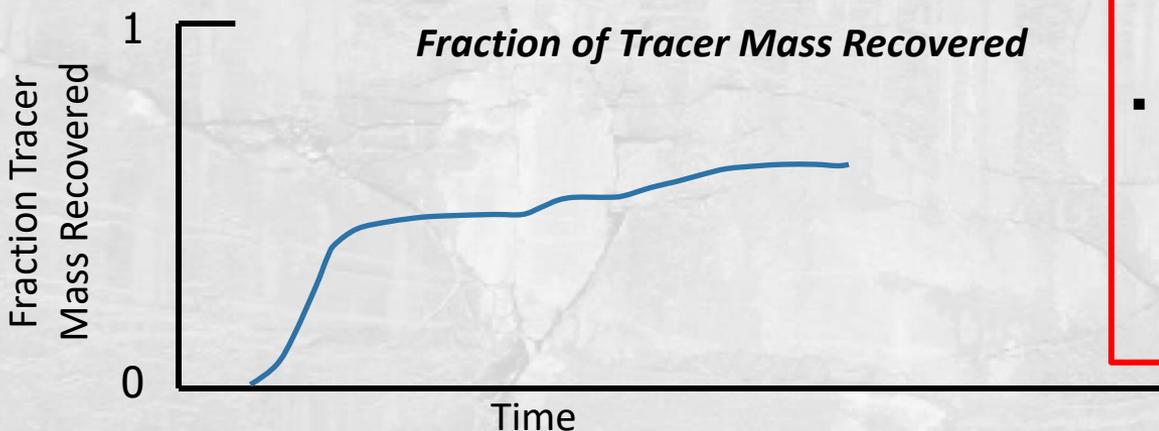
- . . .in fractured rock, the flow regime is unlikely to behave as in a homogeneous porous media. . .
- . . .we often make simplifying assumptions about flow regime to interpret tracer breakthrough curves at pumped well. . .
- . . .can interpretations from controlled hydraulic test be representative of conditions affecting fate and transport of contaminants of interest ?

# Interpreting tracer tests

*Tracer Breakthrough Curve*



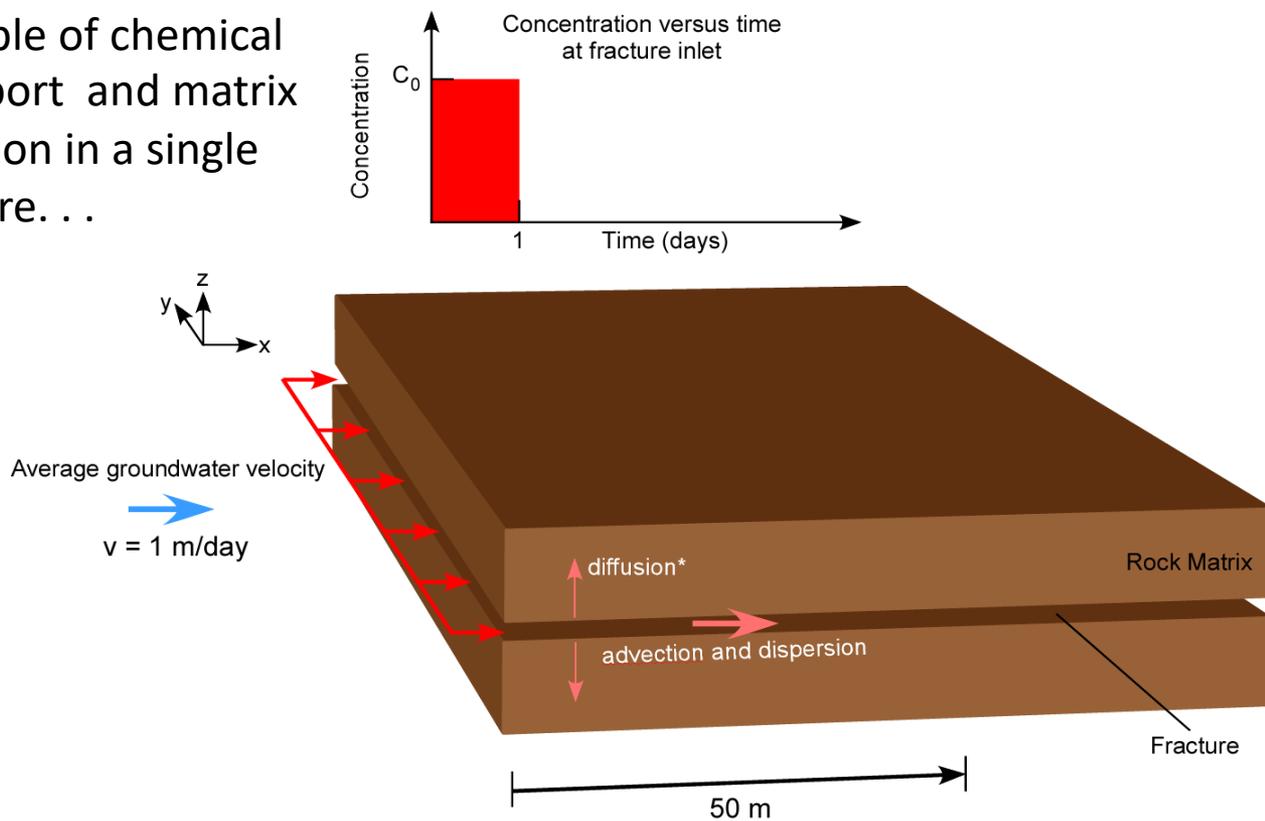
*Fraction of Tracer Mass Recovered*



- Define tracer test objectives to test hypotheses of Conceptual Site Model
- Select tracers to test hypotheses, e.g., chemical dilution, residence time, fracture porosity, matrix diffusion, sorption/desorption, etc.,

# Interpreting tracer tests

Example of chemical transport and matrix diffusion in a single fracture. . .



Dispersion:  $D = \alpha_L |v|$

Matrix diffusion:  $D_m = n \gamma D_w$

\*direction of diffusion depends on the local concentration gradient in the z-direction

Longitudinal dispersivity:  $\alpha_L$

Matrix porosity:  $n$

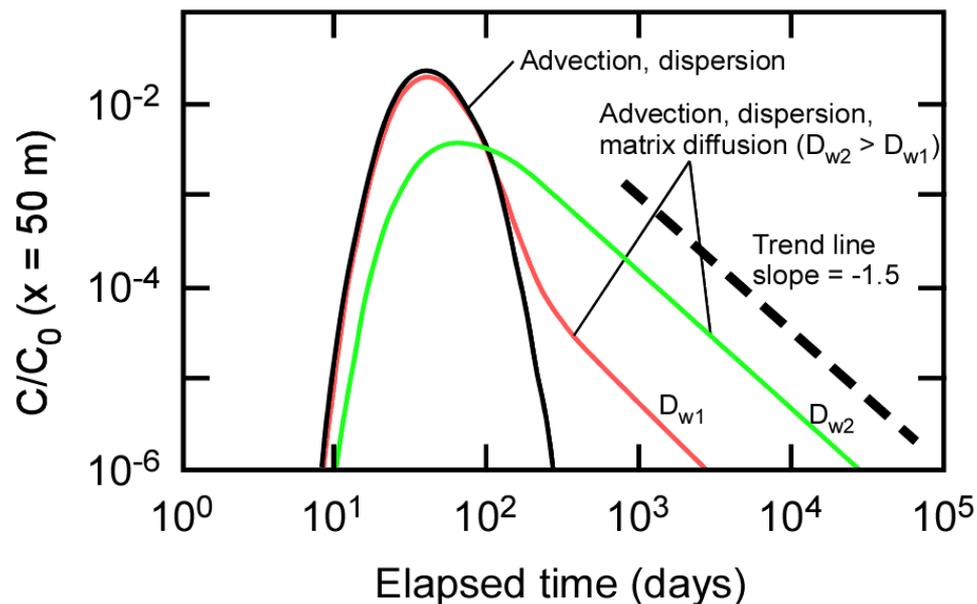
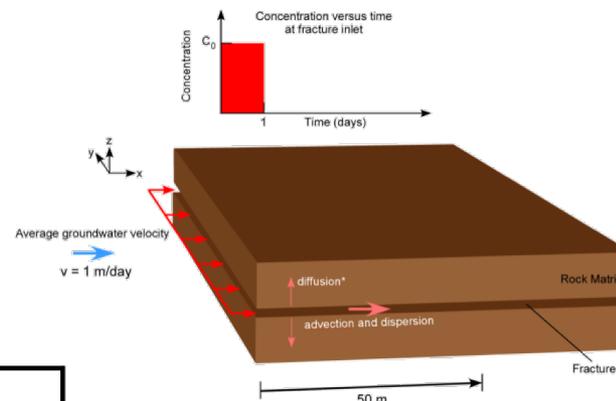
Matrix formation factor:  $\gamma$

Free water diffusion:  $D_w$

# Interpreting tracer tests

Example of chemical transport and matrix diffusion in a single fracture. . .

Chemical response in the fracture (50 m downgradient). . .



Dispersion:  $D = \alpha_L |v|$

Matrix Diffusion:  $D_m = n \gamma D_w$

Longitudinal dispersivity:  $\alpha_L$

Matrix porosity:  $n$

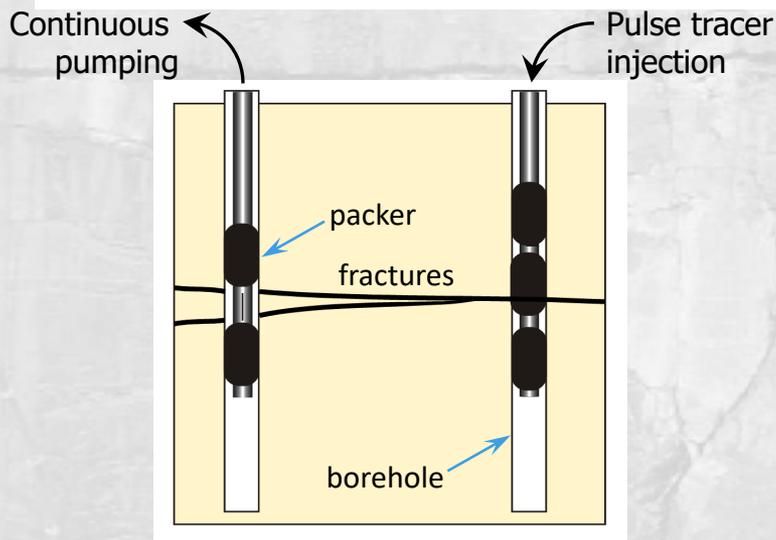
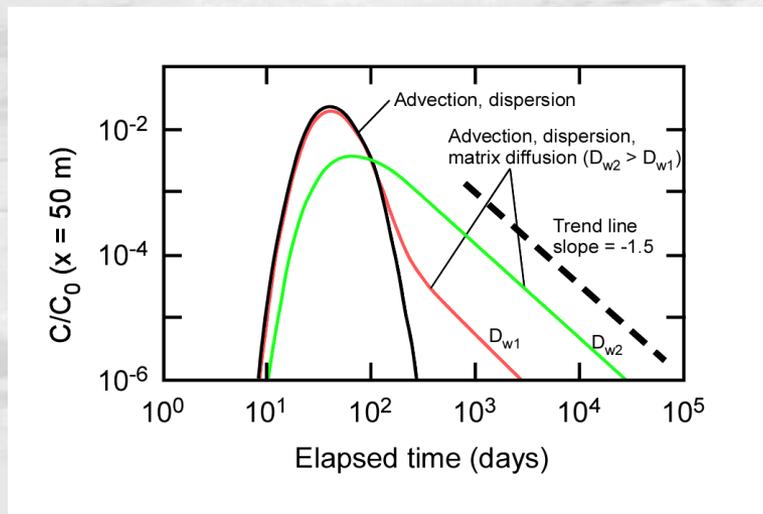
Matrix formation factor:  $\alpha$

Free-water diffusion:  $D_{w1}, D_{w2}$

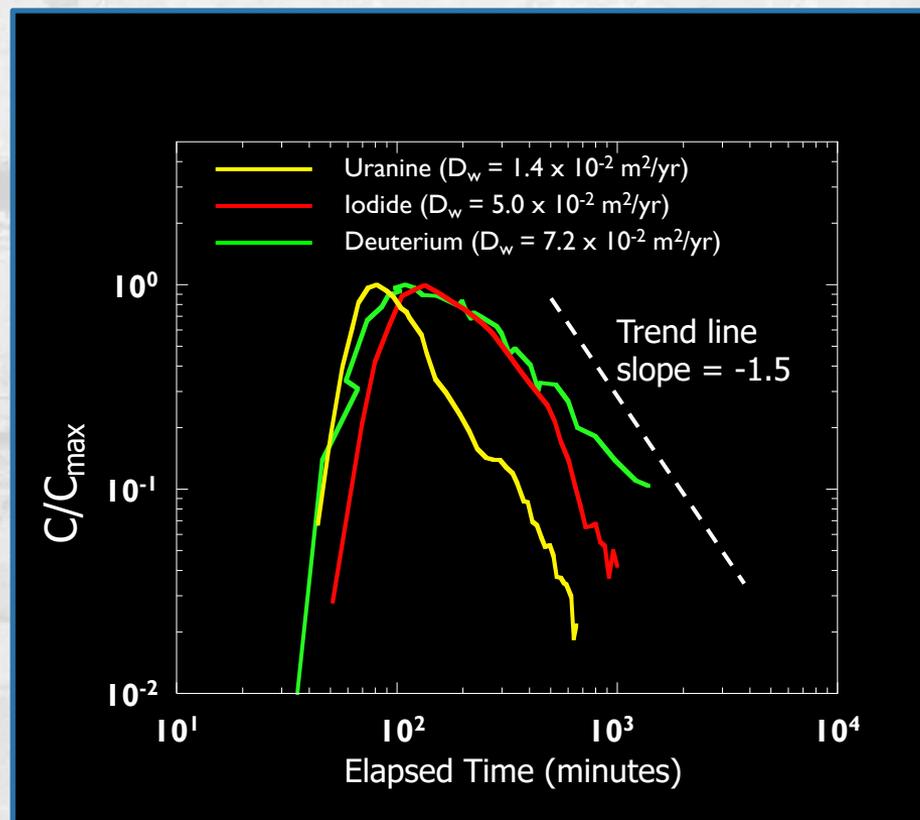
*Shapiro et al., 2007*

# Quantifying Matrix Diffusion – Field Scale Testing (10's of meters)

Simulation – transport in a single fracture



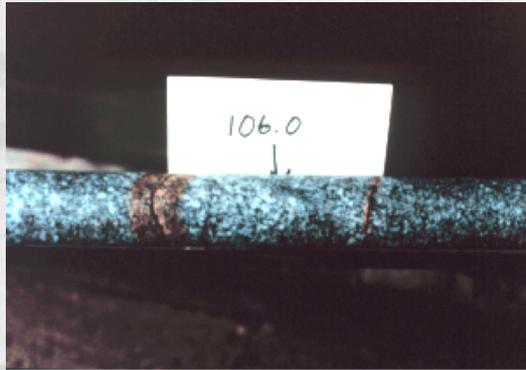
Field Experiment:  
Multiple tracers injected with different  
free water diffusion coefficients,  $D_w$



Chalk Aquifer, Béthune, France, modified from Garnier et al. 1985

# Quantifying Matrix Diffusion – Laboratory Analysis of Rock Core

Diffusion through a rock face



$$J_x = -\frac{n\gamma D_w}{R} \frac{dC}{dx}$$

$J_x$  - diffusive mass flux in the x-direction per unit area ( $ML^{-2}T^{-1}$ )

$n$  - porosity

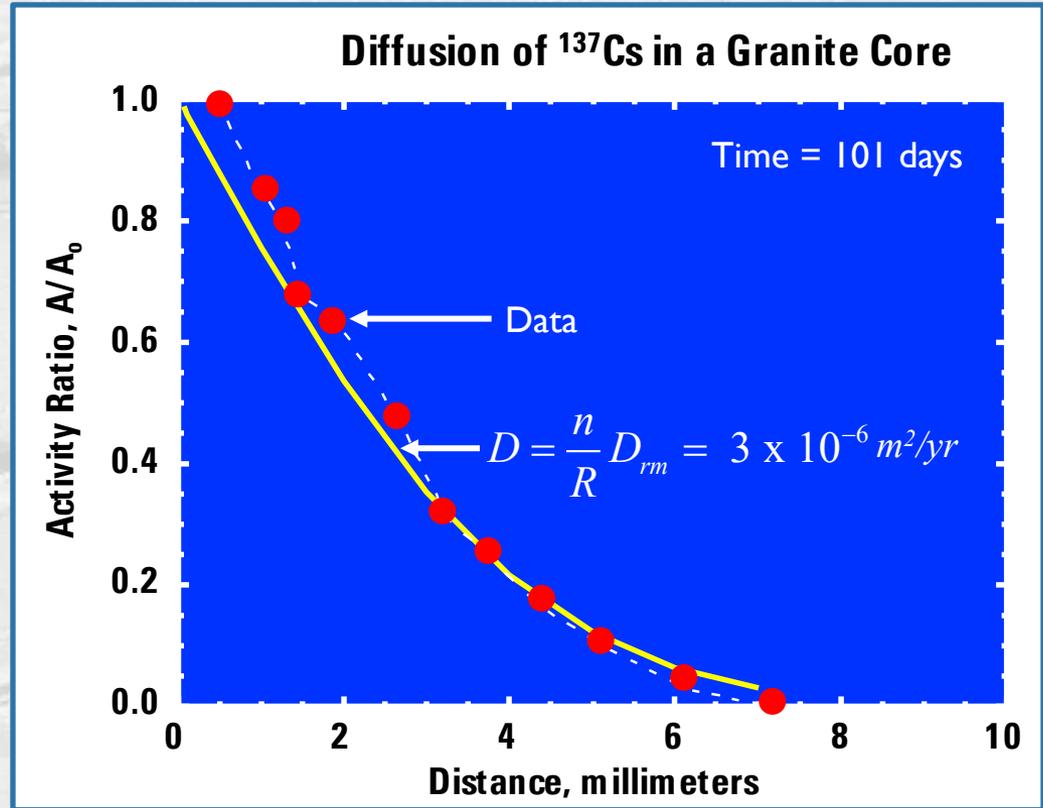
$\gamma$  - formation factor (inversely proportional to tortuosity)

$D_w$  -  $^{137}Cs$  free water diffusion coefficient ( $L^2T^{-1}$ )

$R$  -  $^{137}Cs$  retardation factor

$C$  - concentration (mass per unit volume,  $ML^{-3}$ )

$x$  - spatial coordinate (L)



$$D = \frac{n}{R} \gamma D_w = \frac{n}{R} D_{rm}$$

Are laboratory interpretations appropriate for field scale characterization?

# Quantifying Matrix Diffusion – Field Scale Testing (10's of meters)



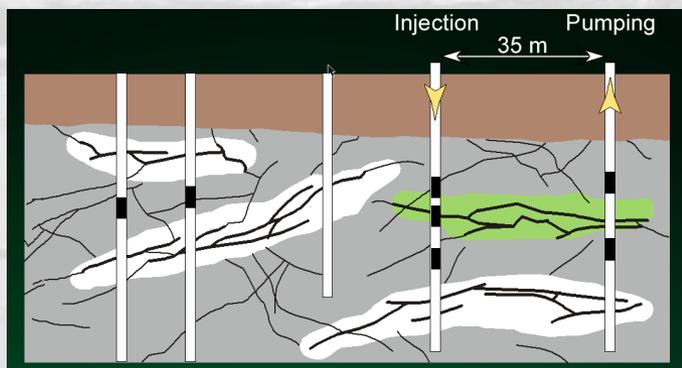
Tracer Injection

Pumping

Not all field scale tests support use of laboratory interpretations to characterize matrix diffusion at the field scale. . .

Granite and Schist – Mirror Lake, NH

Multiple tracers injected with different free water diffusion coefficients,  $D_w$

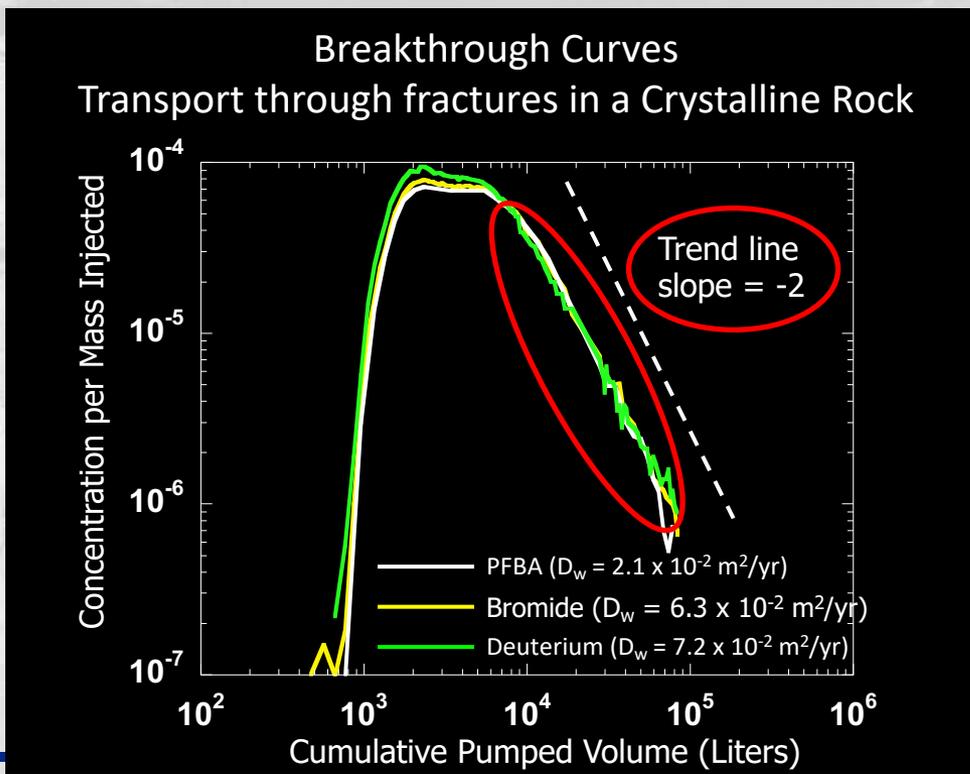


$$D = n\alpha D_w = nD_{rm}$$

However, from interpretation of breakthrough curves:

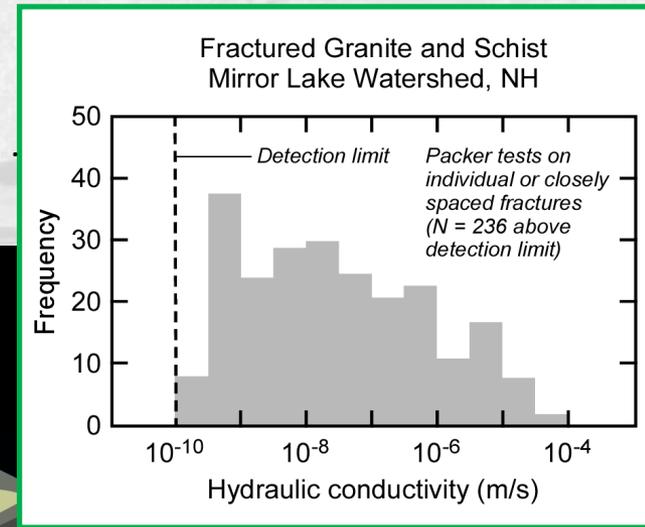
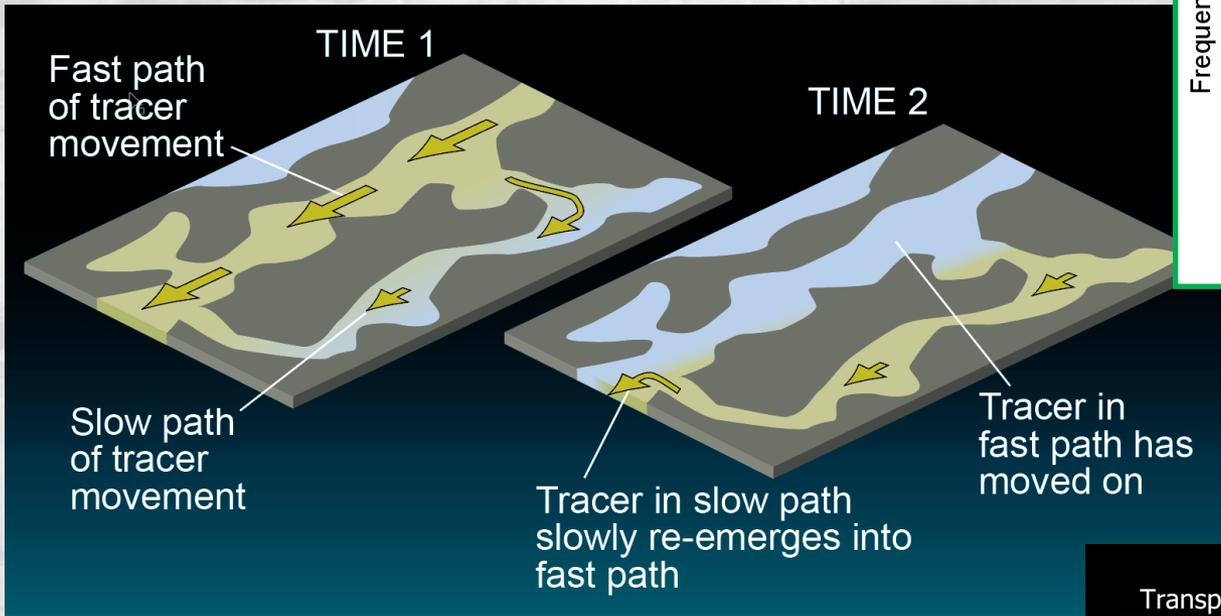
$$D_{rm} > D_w$$

Is this physically reasonable?

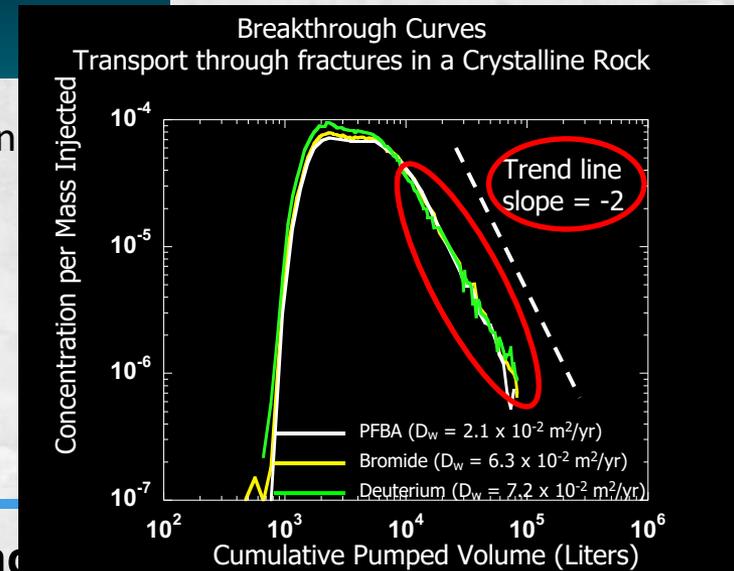


# “Effective” Matrix Diffusion

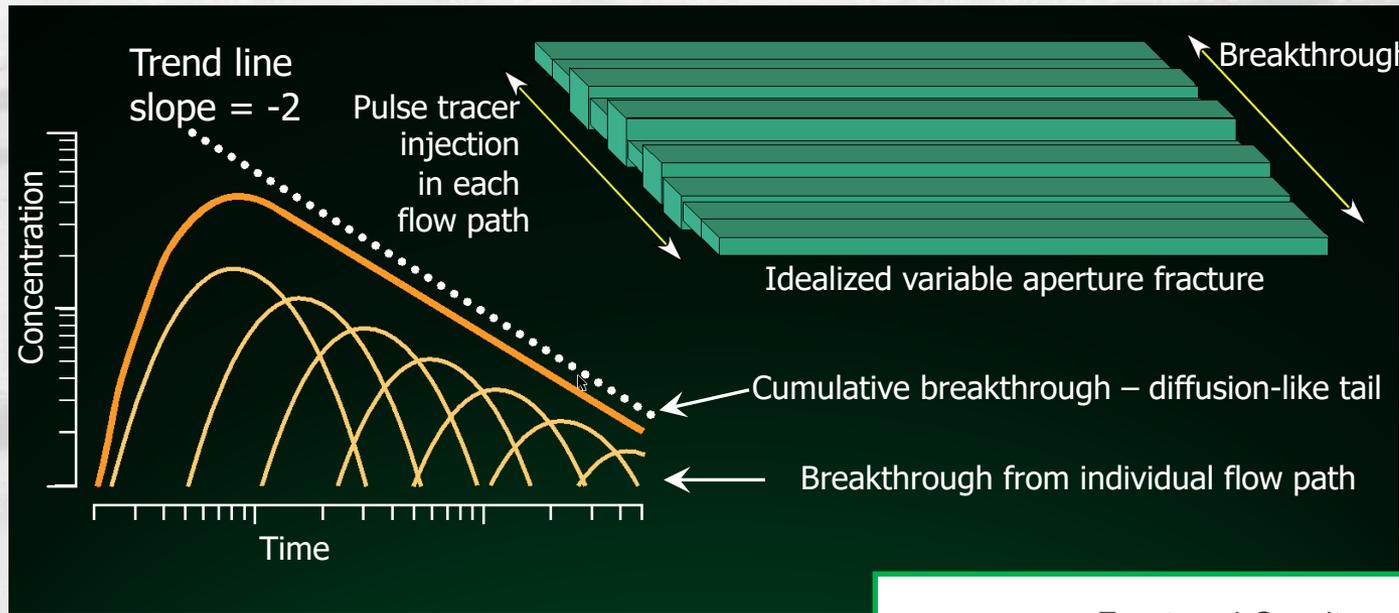
Diffusion-like tailing ( $D_{rm} > D_w$ ), leading to extended residence times. . .  
 An artifact of “extreme” variability in fluid advection



- Hydraulic conductivity of fractures varies over more than 6 orders of magnitude
- Large range of velocity is not consistent with the conceptualization of hydrodynamic dispersion
- Slow advection leads to a diffusion-like process – small travel distance over extended period time, similar to diffusion into and out of flow-limited aquifer material



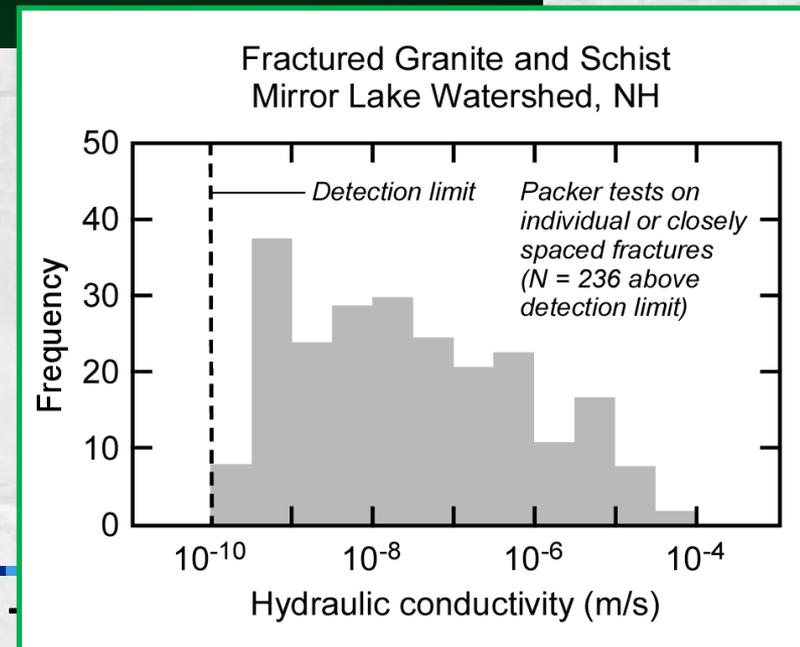
# “Effective” Matrix Diffusion

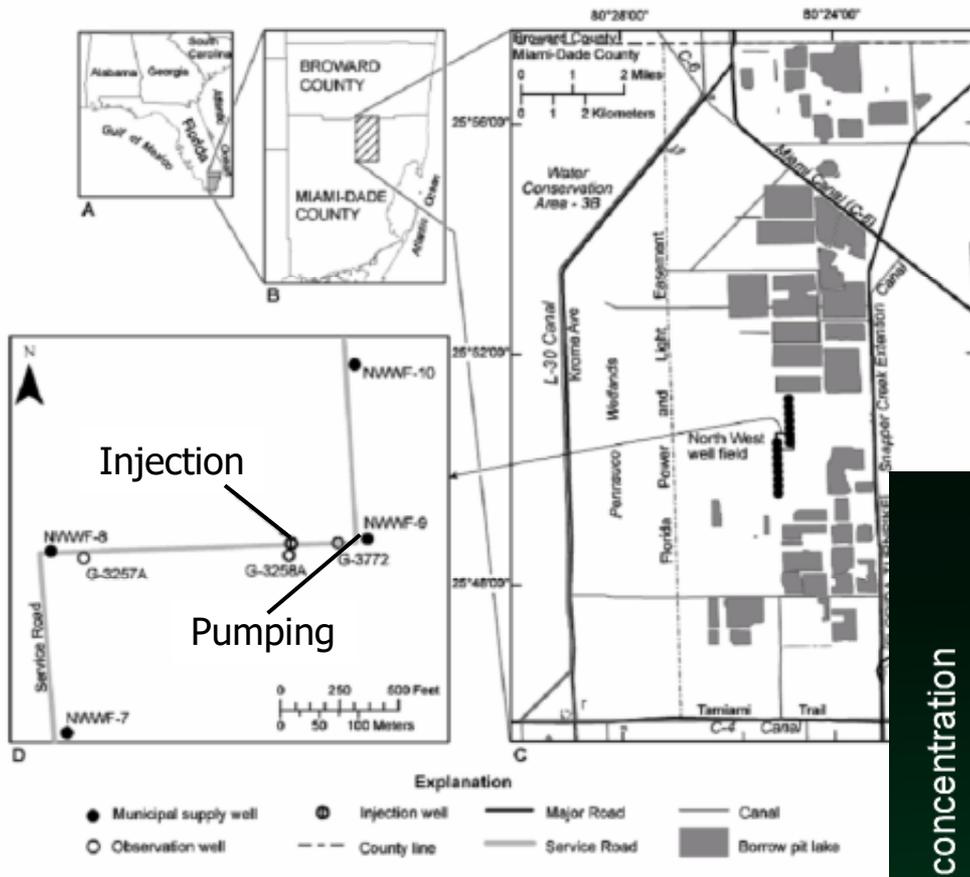


“Effective” diffusion impacts calculations of the longevity of contamination. . .

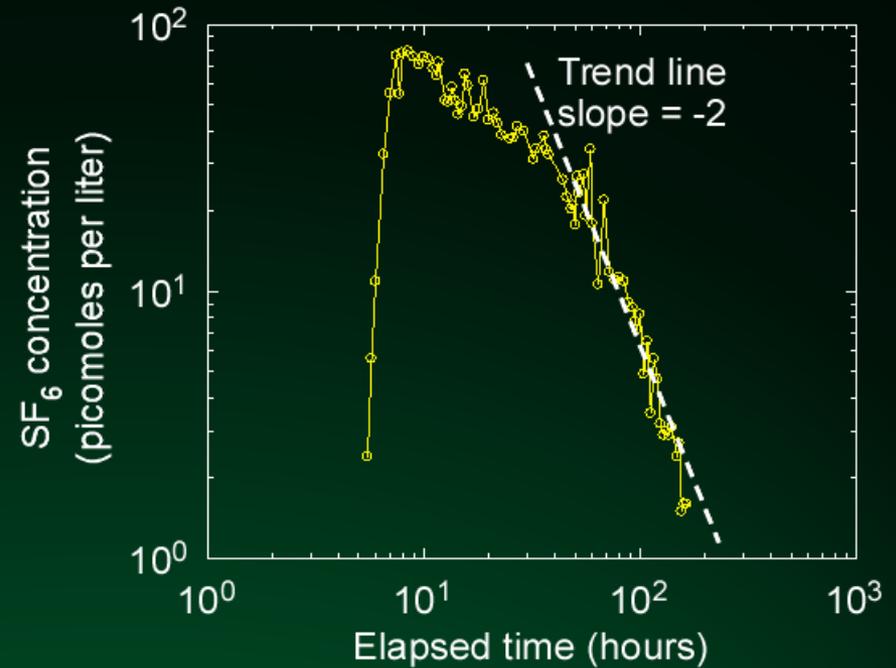
. . .contaminant storage and release exists not only in the rock matrix. . .

. . .but also, in low-permeability fractures. . .

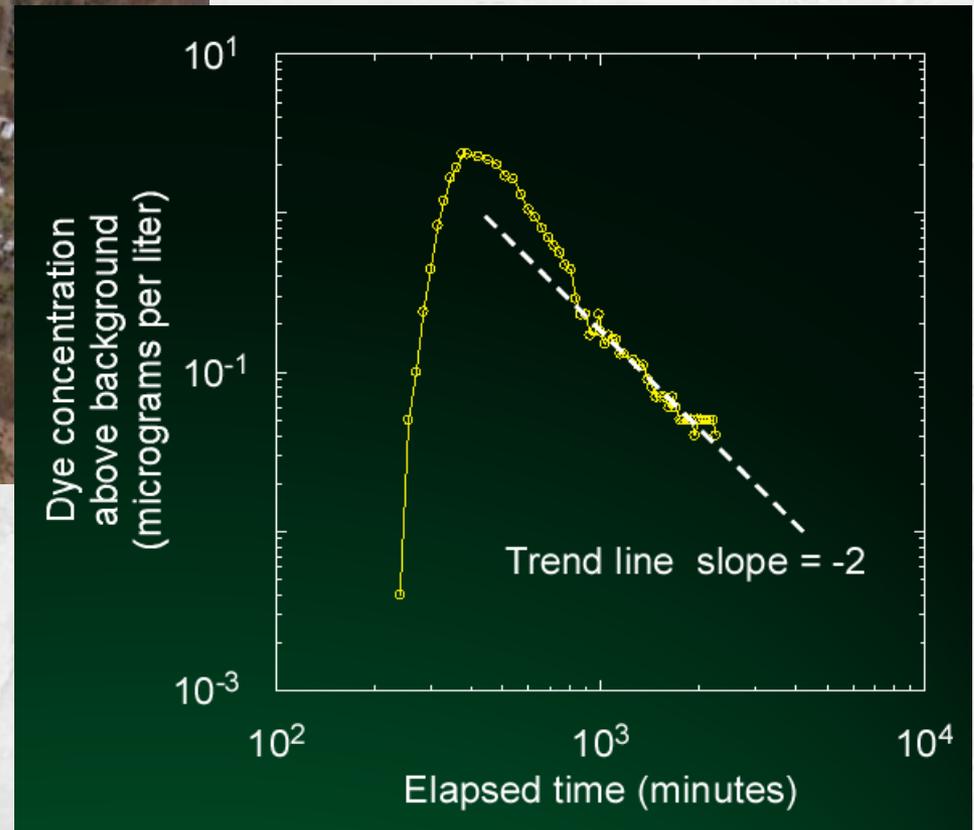
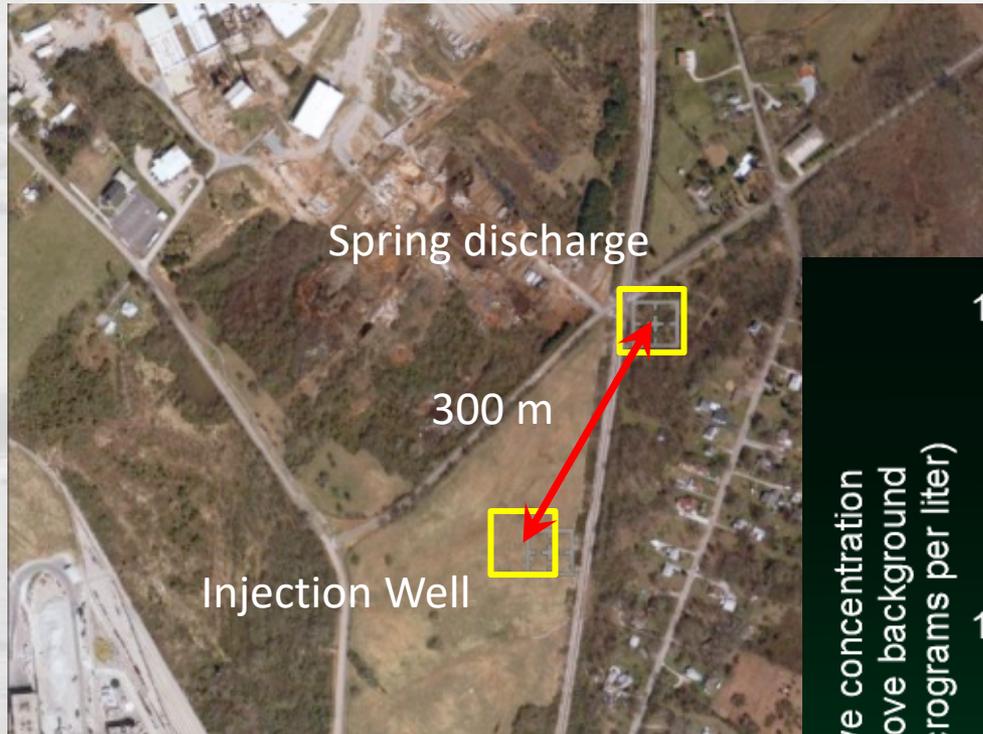




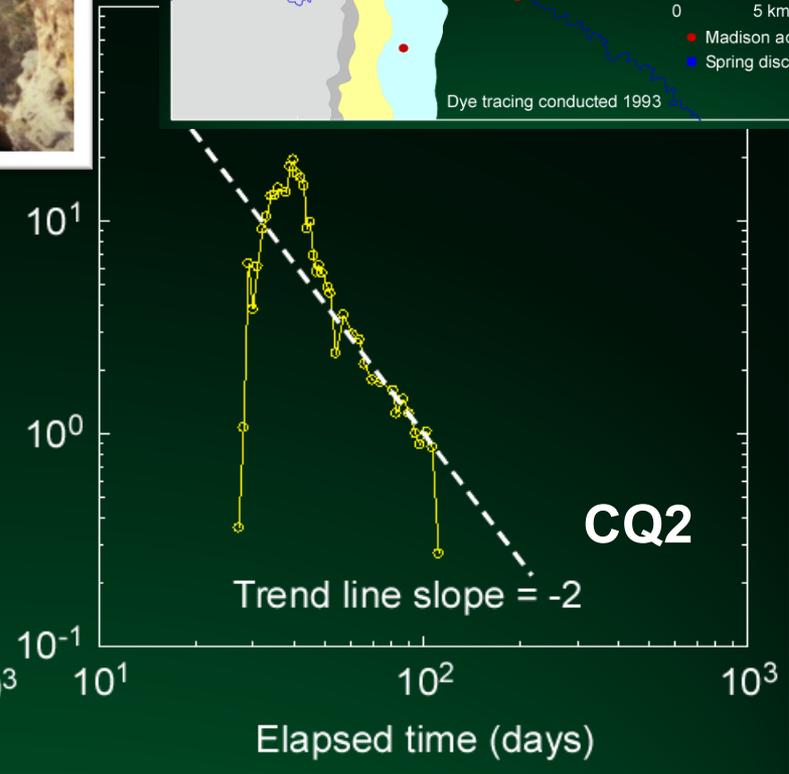
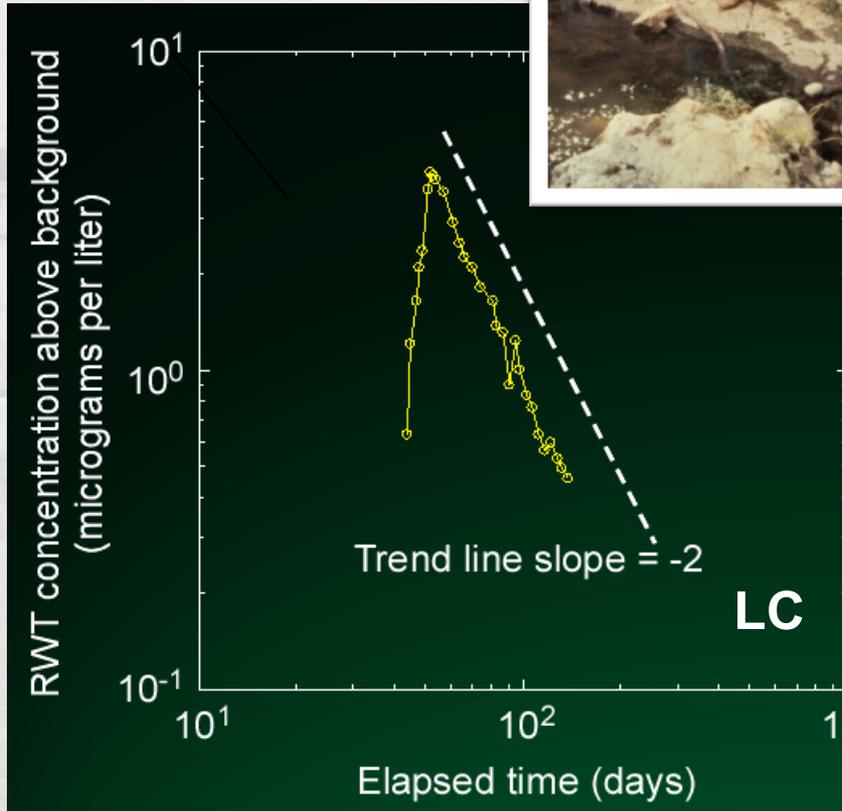
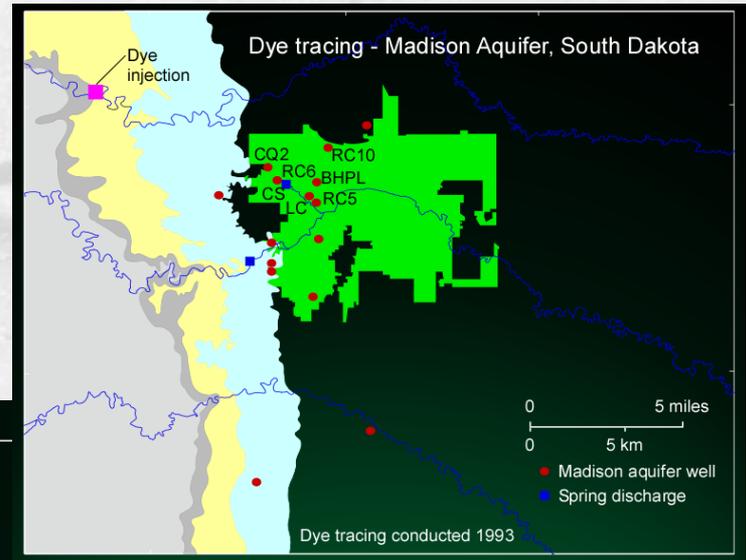
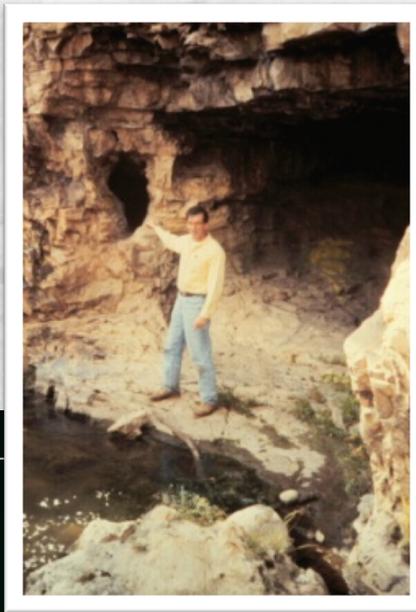
Biscayne Aquifer, Miami  
Limestone - ~100 m



# Maryville Limestone, Alcoa, Tennessee - ~300 m



# Madison Limestone Rapid City, SD ~10 km



# Final Thoughts

- ❑ Tracer tests can provide a direct measure of fluid velocity, chemical dilution and attenuation processes, chemical and biological reactions associated with groundwater contaminants
- ❑ Tracer tests provide valuable information to (1) conceptualize processes affecting fate and transport of contaminants, (2) design and implement remediation strategies
- ❑ Tracer tests in fractured rock are likely to be successful under hydraulically stressed conditions, provided the hydraulic perturbation can be monitored in cross-hole tests
- ❑ Single-hole tracer tests are used primarily to estimate *in situ* process and identify local parameter values. . .less reliable in estimating groundwater velocity

# Selected References

(Note: This is not an exhaustive list. Please contact [ashapiro@usgs.gov](mailto:ashapiro@usgs.gov) for a more extensive list of references.)

- Becker, M. W. and Shapiro, A. M. 2000. Tracer transport in fractured crystalline rock: Evidence of nondiffusive breakthrough tailing. **Water Resources Research** 36(7): 1677-1686. 10.1029/2000WR900080.
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