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Colloid-Facilitated Transport: Studies Related to CFM Project at GTS

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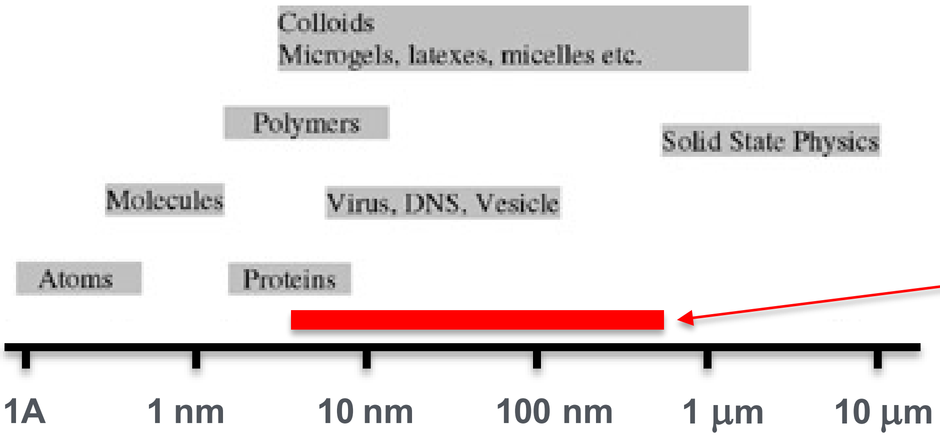
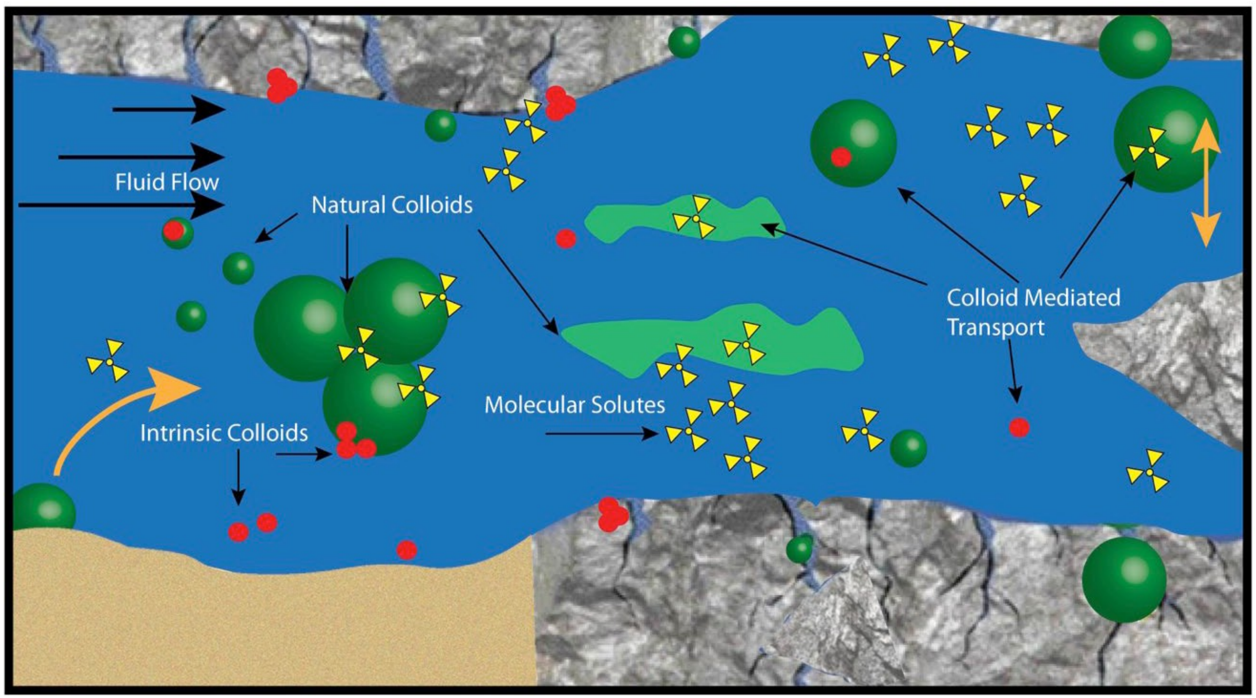
Colloid-Facilitated Transport (CFT) of Radionuclides

Colloids

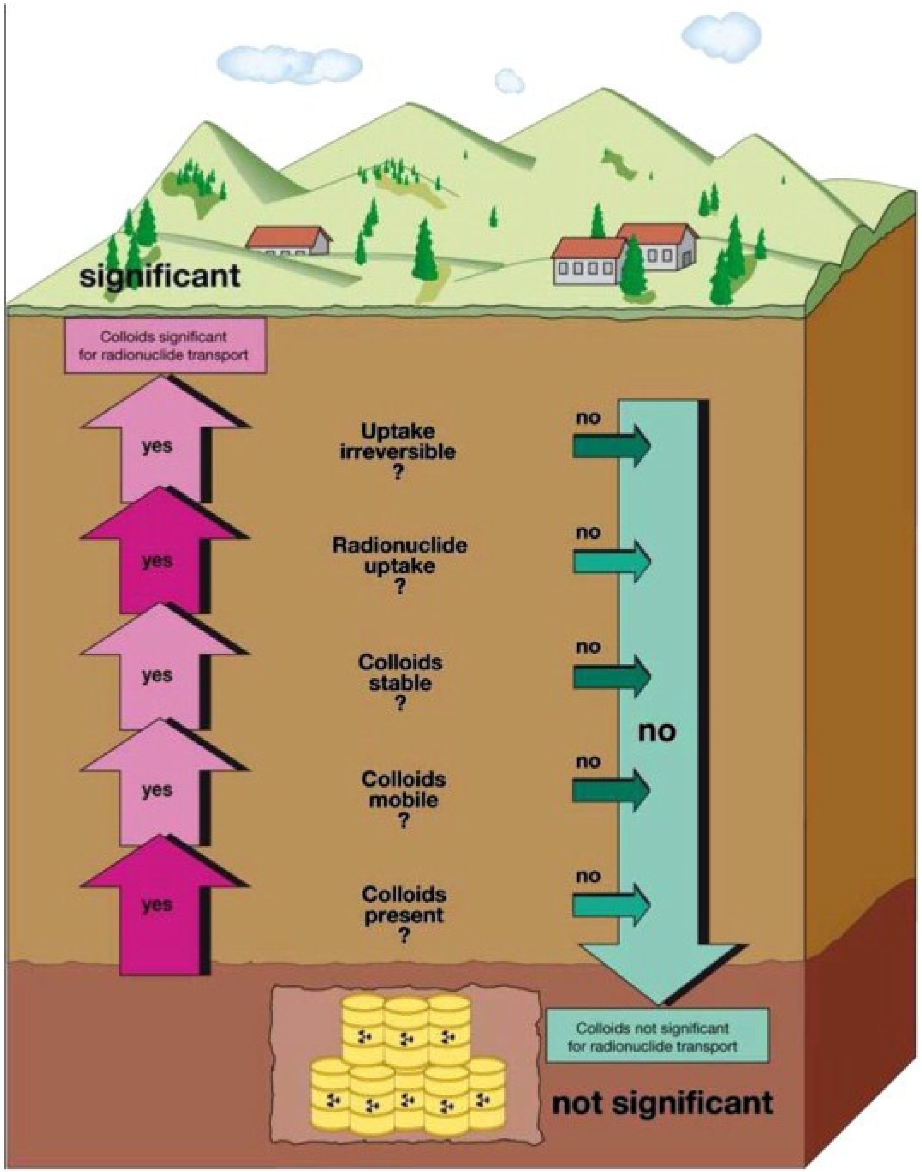


Approximate Practical Size Range of Interest for Colloid Transport over Large Scales in Geologic Media

Colloid-Facilitated Transport of Solutes



The "CFT Ladder"



In Words:

For CFT to be a problem, you need stable colloids that are capable of migrating long distances,
AND you need radionuclides to be very strongly associated with these colloids

CFM (Colloids Formation and Migration) Project Overview

Structure of the CFM Project

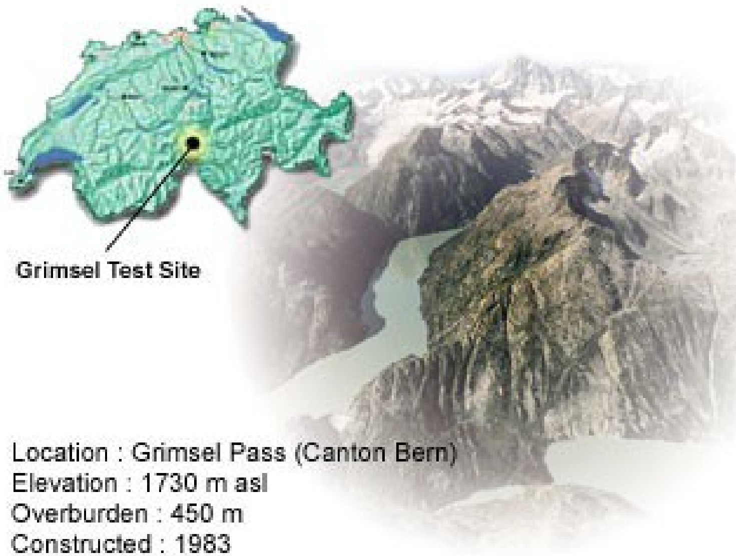
Laboratory studies	**Field experiments**	**Modelling studies**
Colloid-Rn interaction Colloid Generation Field test analysis	In situ test: formation & Migration tests with colloids, homologues, Rn tracers	Solute, colloid and associated Rn transport Colloid generation

- •Colloid generation
- •Colloid transport/retardation and stability
- •Radionuclide association
- •Bentonite intercomparison (MX-80, Febex, Kunigel)

- •Site characterization and site preparation
- •Assessing the advective travel times
- •Analyzing the recovered tracer mass
- •Estimating dispersion parameters in the shear zone flow fields

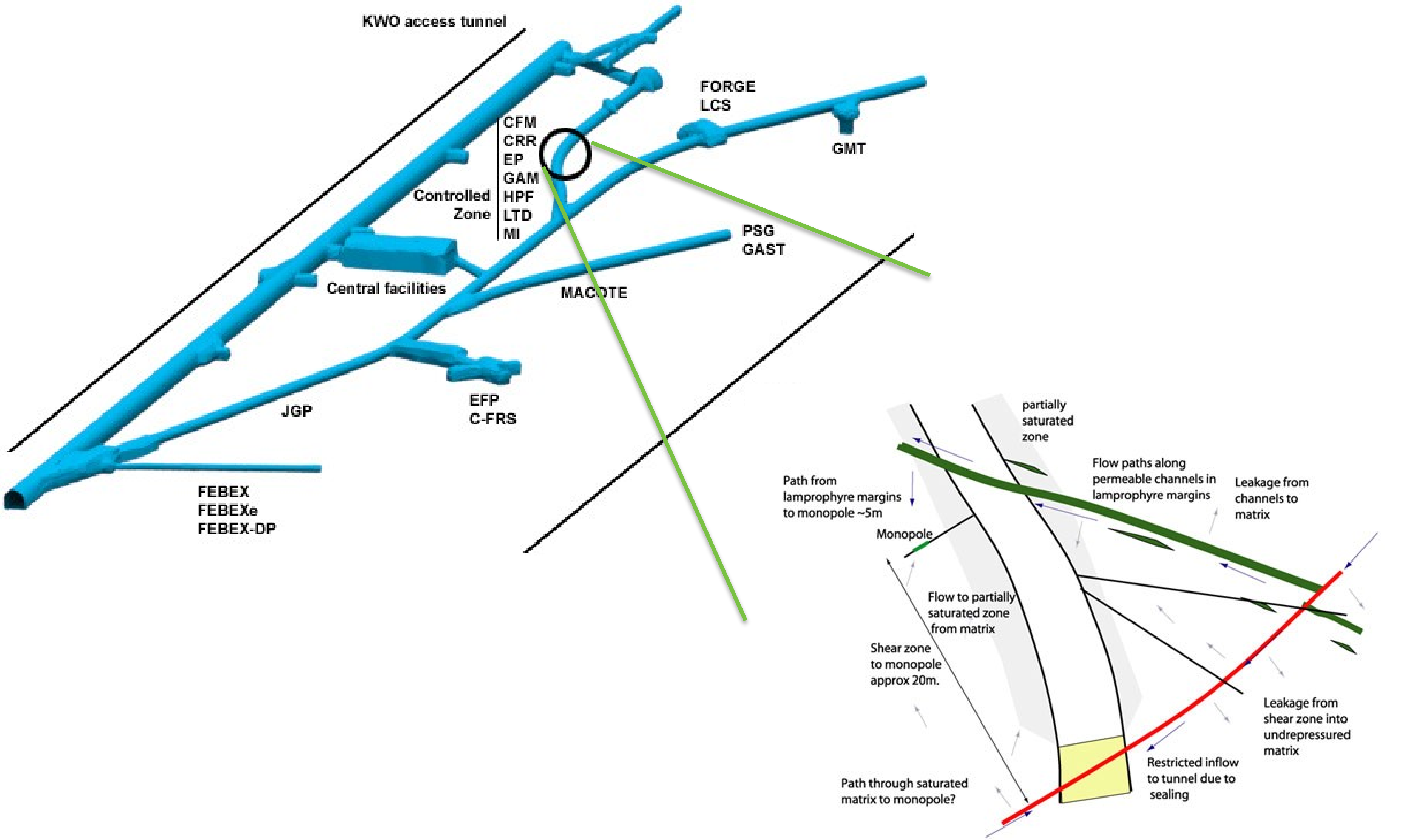
- •Supporting the in-situ tests
- •Initiating performance-assessment relevant studies on colloid generation and on colloid-facilitated radionuclide transport

Grimsel Test Site



(1) Grimsel Test Site, (2) Rätrichsbodensee,
(3) Grimselsee, and (4) Juchlistock

The GTS Underground Facilities



CFM in-situ experiment

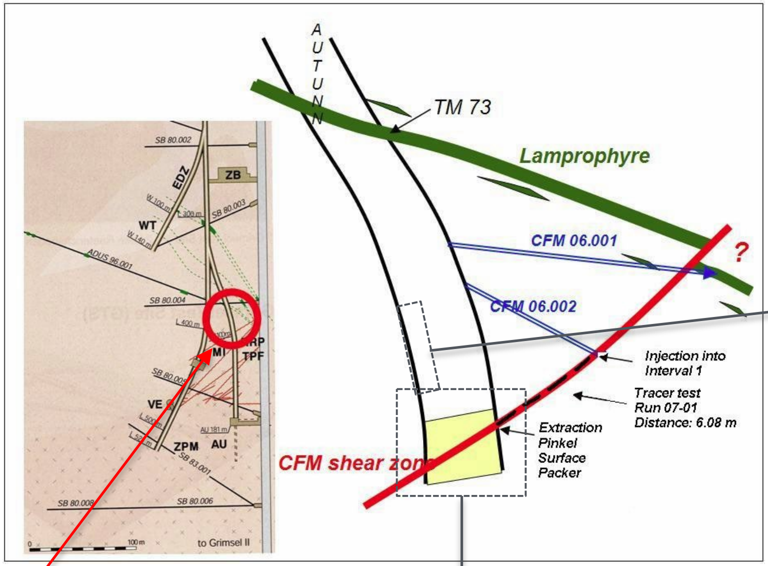
CFM (Colloids Formation and Migration) Project Overview

- CFM began in 2004 (preceded by CRR, Colloid and Radionuclide Retardation Project, 1998-2003)
- U.S. was formal partner in 2013-2015, with informal involvement since 2006
- Focus has always been on bentonite colloids in fractured crystalline media (a granodiorite at GTS)
  - Relevant Scenario: Waste package breach allows radionuclides to sorb onto bentonite backfill, which subsequently erodes into flowing fractures, carrying radionuclides away on colloids
- Distance Scales: ~2 – 6 meters
- Time Scales: 1 – 60 hours (mean residence times), with general progression of increasing time scales
- RN-doped bentonite plug emplacement in 2015

CFM Project Testbed

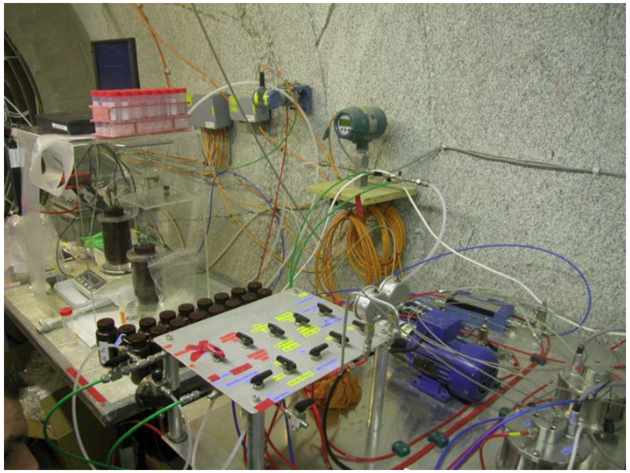
Hydraulic Isolation and Control of Shear Zone Inflow via 3.5-m Diameter "Packer"

Plan View of Testing Area

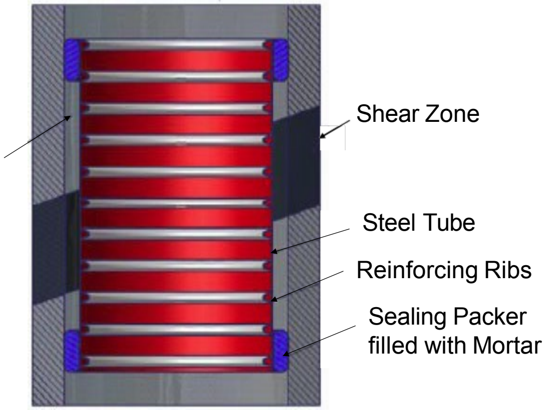


Radiological Control Area

Testing Control and Data Acquisition System

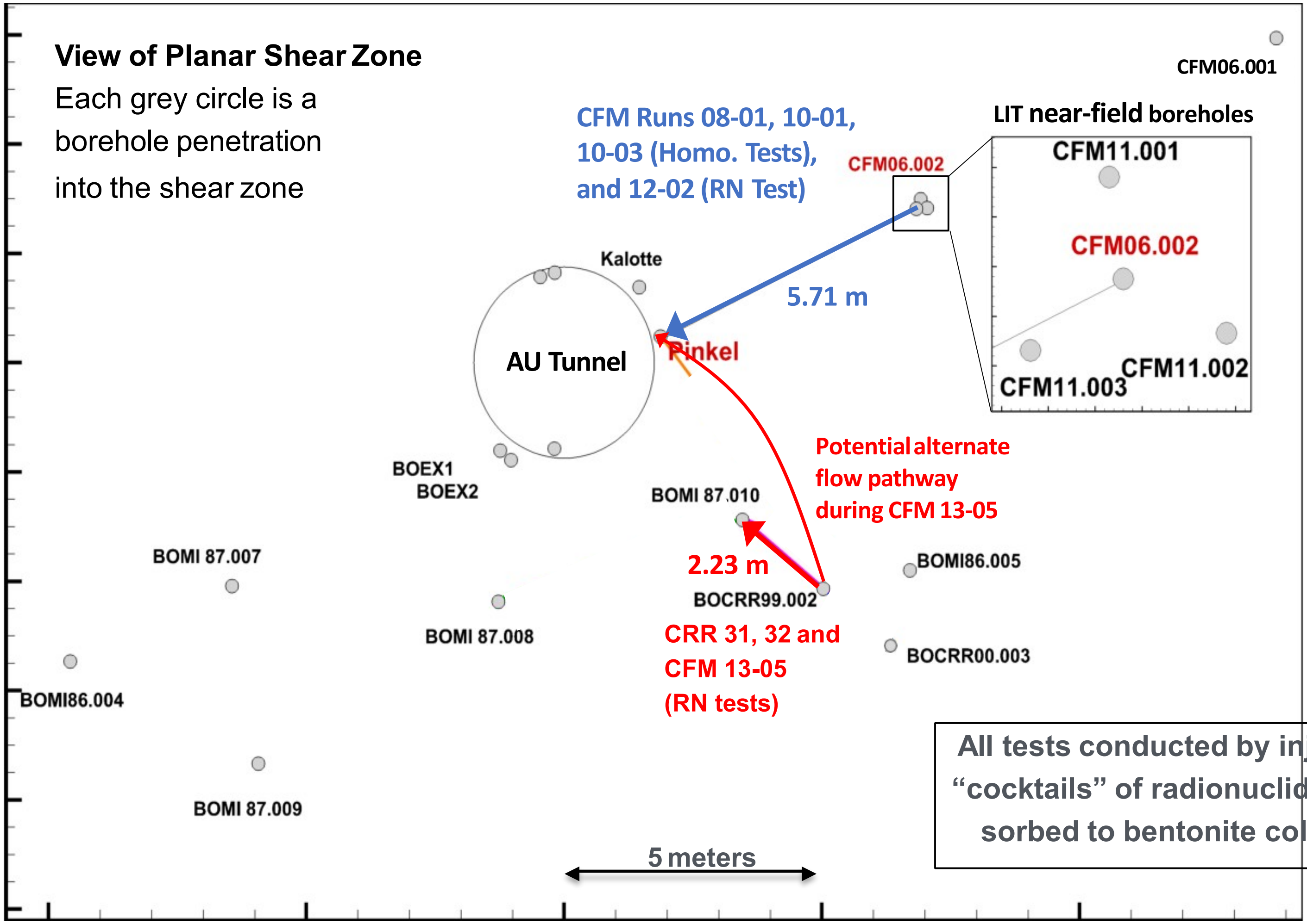


Pinkel Surface Packer



Colloid-Facilitated Transport Tests (2002-2013)

6 CFT Tests: 3 with tri- and tetravalent "homologues", and 3 with radionuclides (also one radionuclide test without colloids, CRR 31)



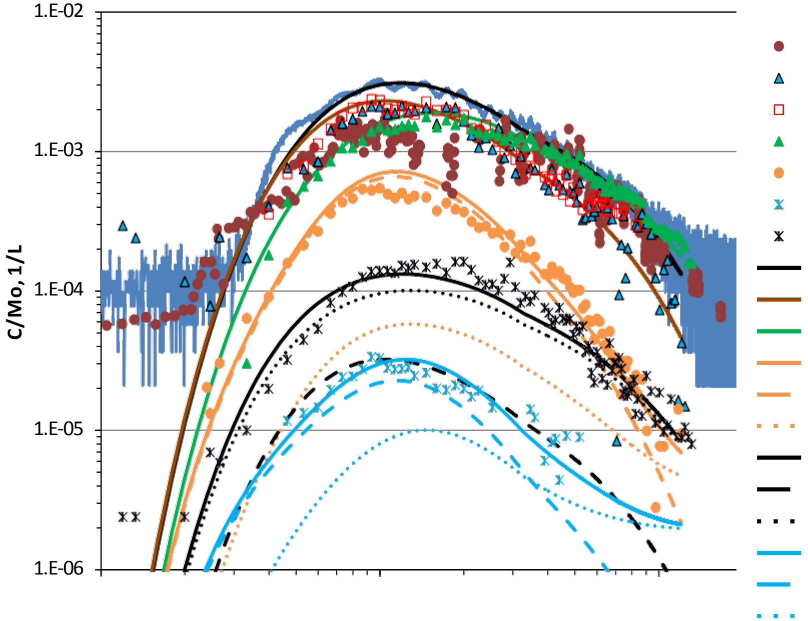
All tests conducted by injecting "cocktails" of radionuclides pre-sorbed to bentonite colloids

Example of Model Interpretations (CFM Test 12-02)

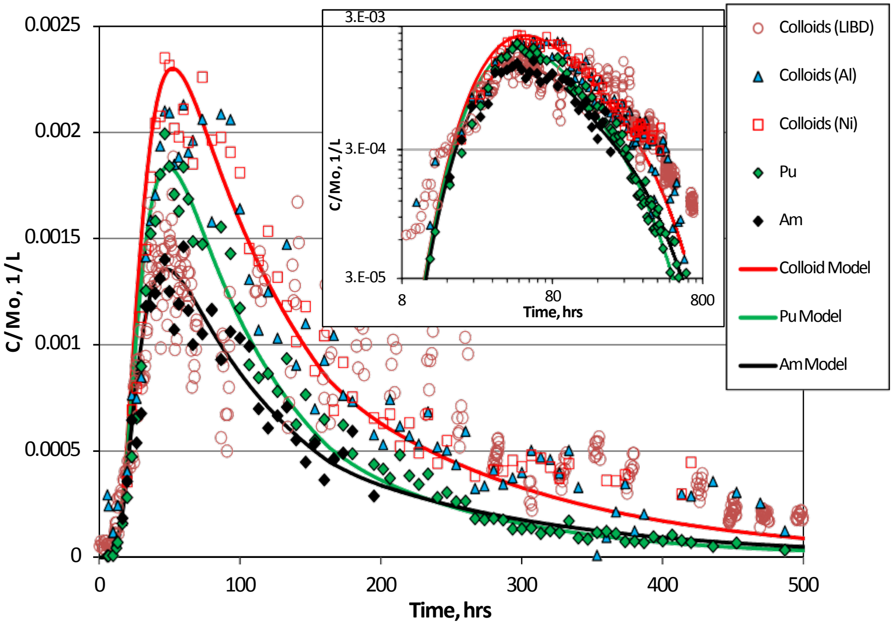
Modeling Approach:

- **Model Conservative Tracer First (Amino-G Acid, or AGA)**
- **Then Model Colloids Using Filtration Parameters Coupled with Conservative Transport**
  - **Account for lower colloid recovery relative to conservative tracer by filtration processes**
- **Then Model Radionuclides using Sorption/Desorption Parameters Coupled with Colloid Transport**
  - **Account for lower radionuclide recovery relative to colloids by RN desorption from colloids**

Conservative Tracer (AGA), Colloids, and Selected Radionuclides

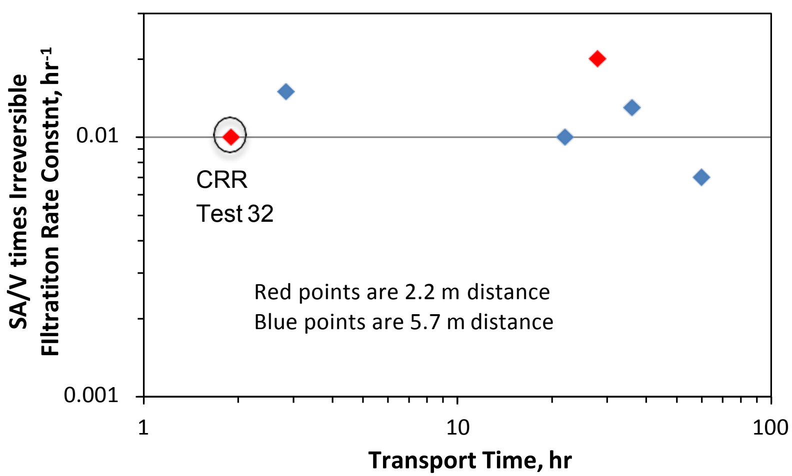


Colloids, Pu, and Am



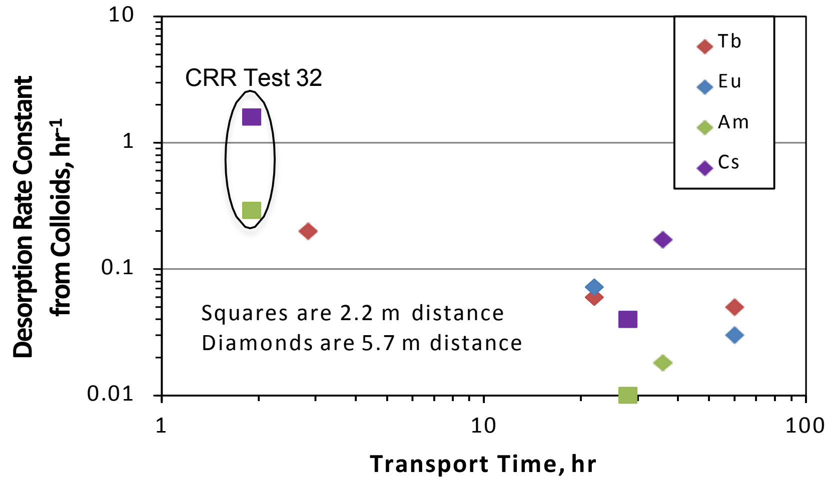
Summary of 2002-2013 Results

Bentonite Colloid Filtration Rate Constants

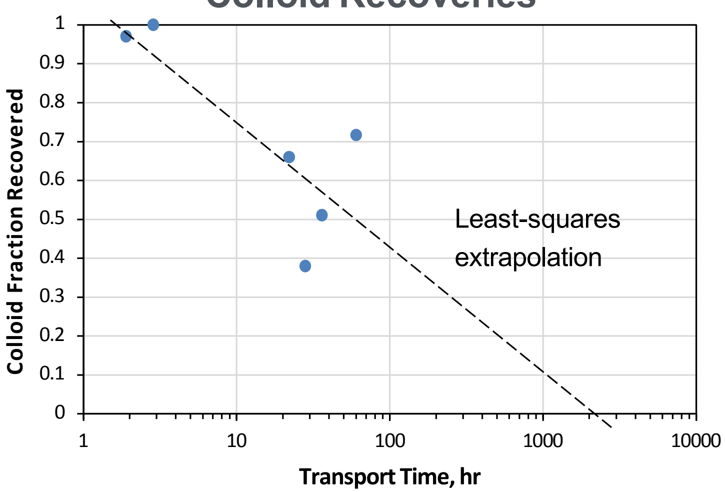


RN/Homologue Desorption Rate Constants

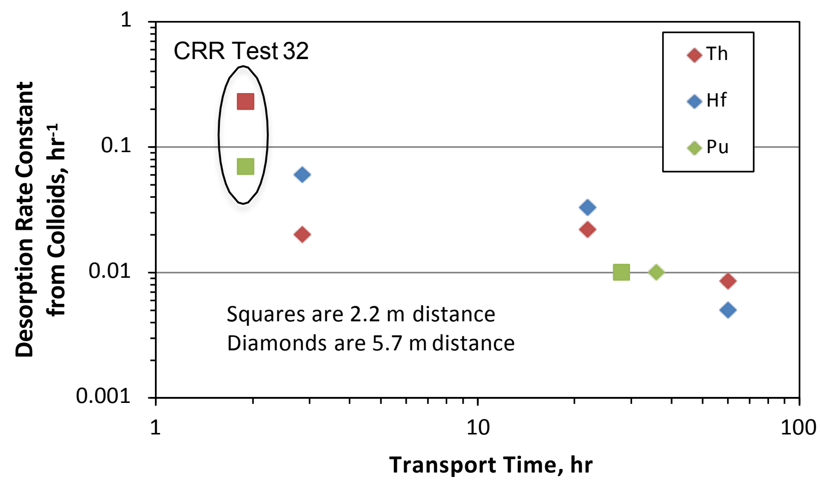
Trivalents and Cesium



Colloid Recoveries

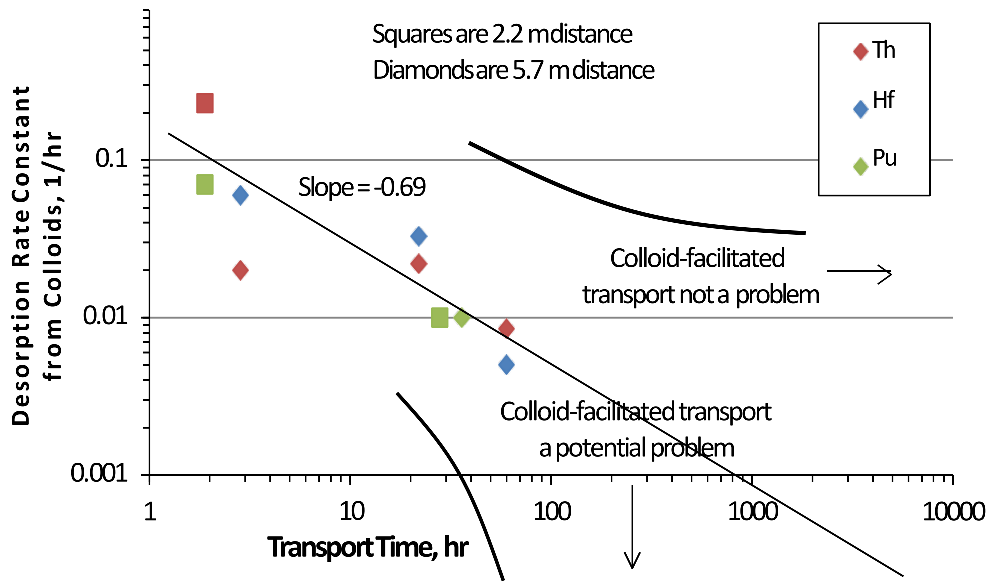


Tetravalents



Upscaling Questions

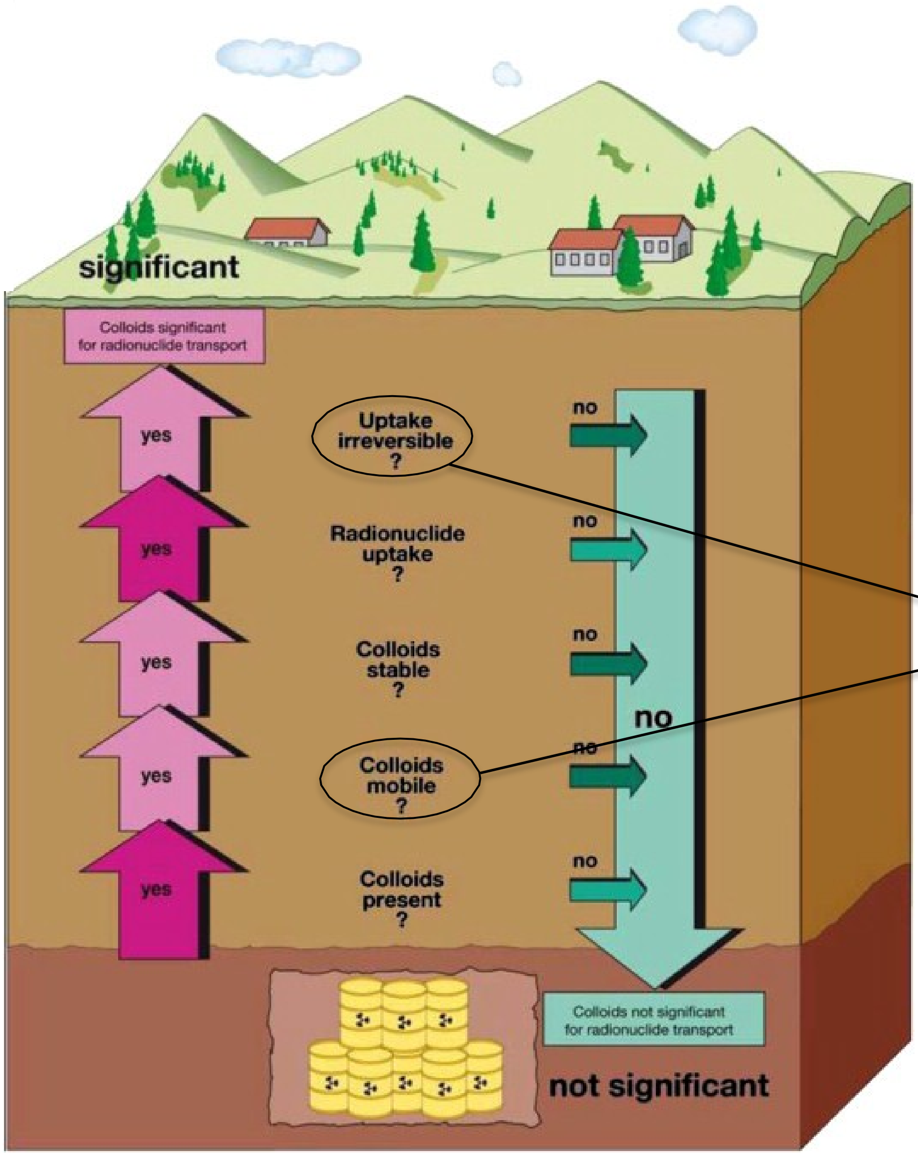
Can a Simple Extrapolation be Applied?



This plot is not interpreted as a literal decrease in desorption rate constants with increasing time scale, but rather as a revelation of stronger and stronger sorption sites (with smaller desorption rate constants) as time scales increase.

The key question is: Are there any sorption sites with slow enough desorption rates to be effectively irreversible over repository time and distance scales?
And if so, are there any colloids that will remain mobile over these time/distance scales?

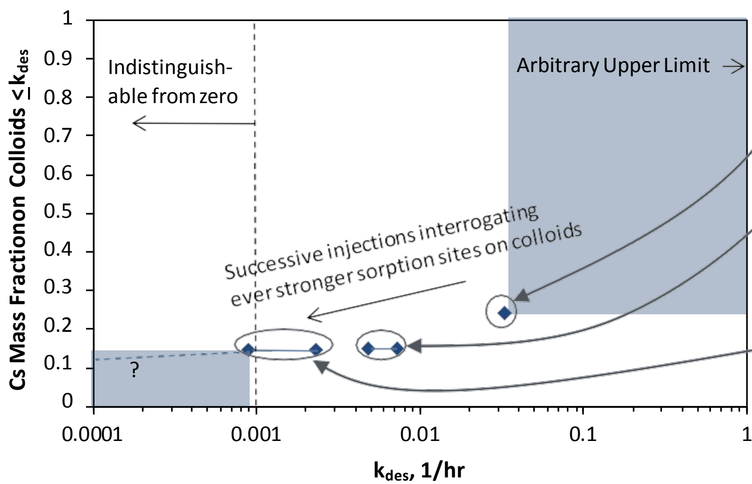
CFM and the "CFT Ladder"



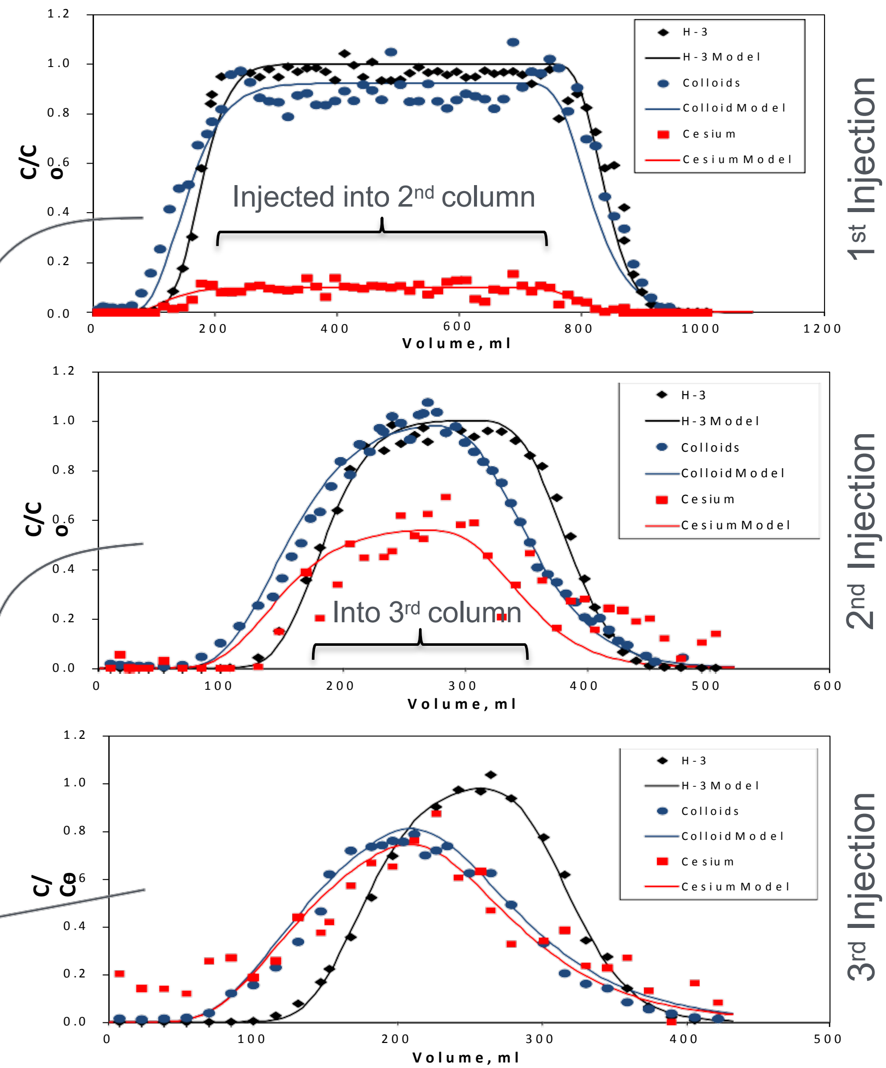
<u>CFM answers</u>:
Yes, for up to 100 hrs and 6 meters in case of bentonite colloids in groundwater with ~0.7 mM ionic strength, but extrapolation to longer time and distance scales is a big uncertainty

Recent Approach in Lab Testing: Cs Associated with NNSS Colloids

Columns

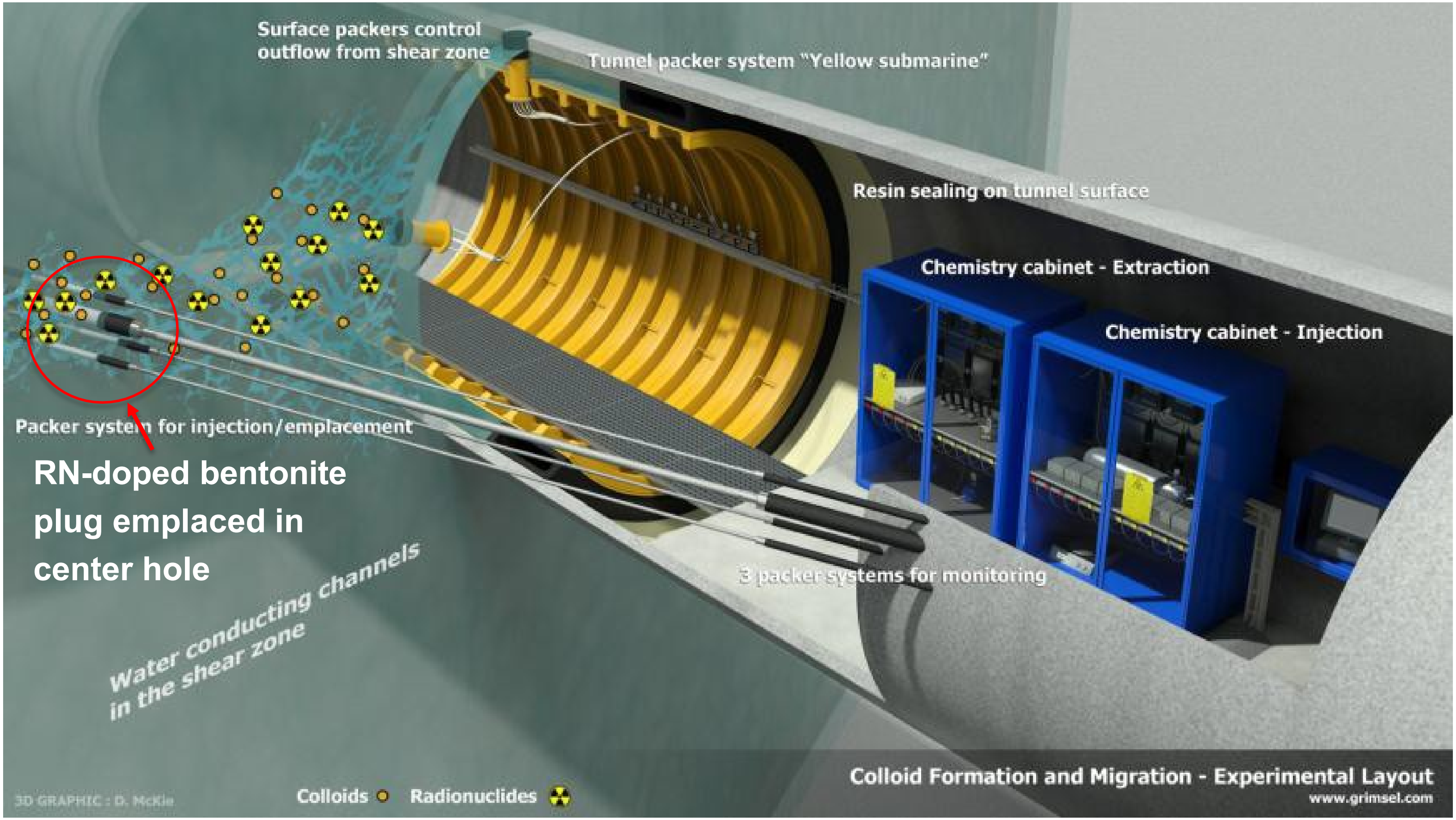


Repeat Injections of Cavity Water (~48-hr Residence Time)



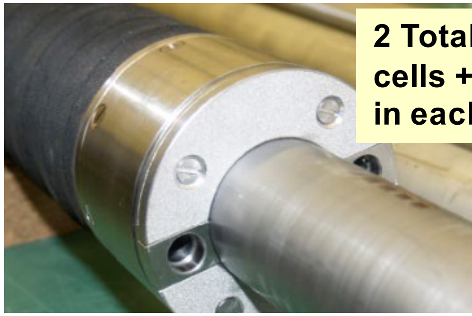
Each successive injection interrogates stronger Cs sorption sites on colloids

CFM Long-Term In-Situ Test (LIT): 2015-present



Shear-zone flow kept the same as in CFM 12-02 test

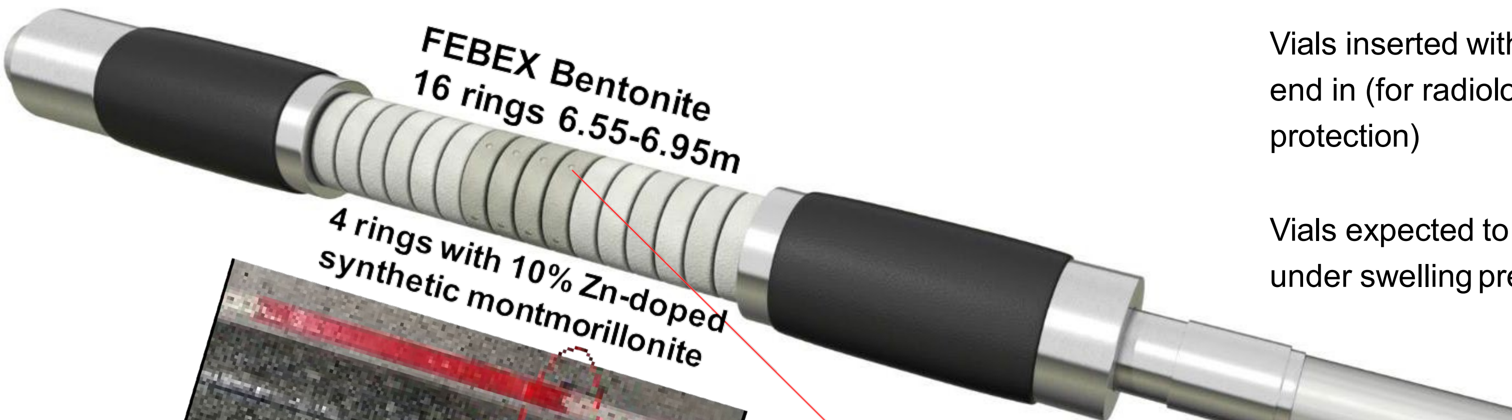
RN-doped bentonite emplacement details

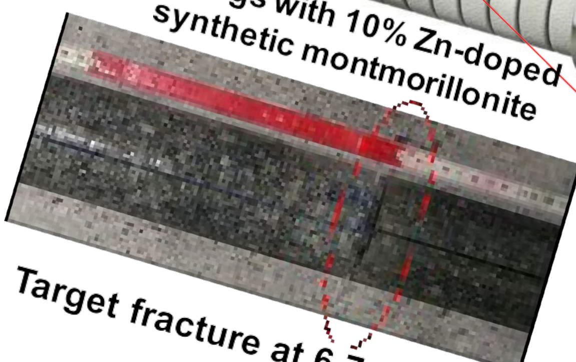


2 Total Pressure cells + piezometer in each packer face

Precompacted rings of FEBEX bentonite:

- Outer diameter: 82mm
- Inner diameter: 43mm
- Dry density: 1.65Mg/m^3
- Gravimetric water content:~14%

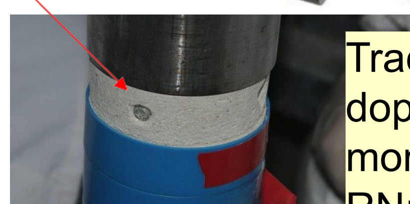




Target fracture at 6.7-6.8m

Vials inserted with open end in (for radiological protection)

Vials expected to break under swelling pressure

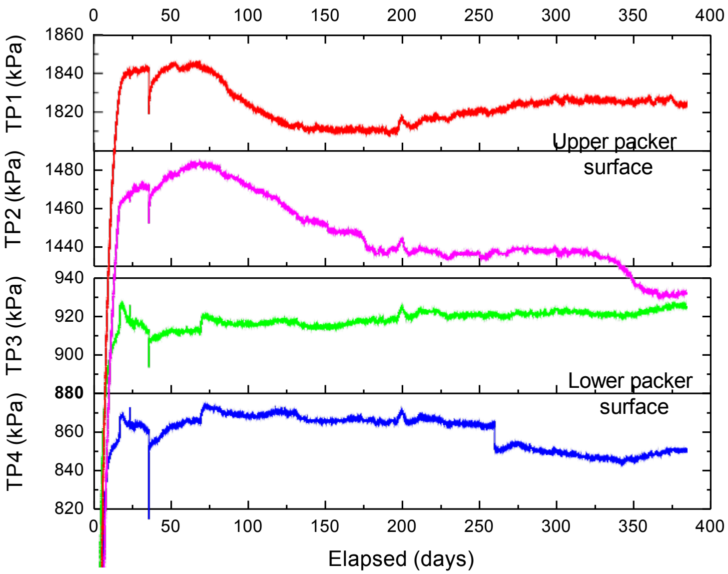


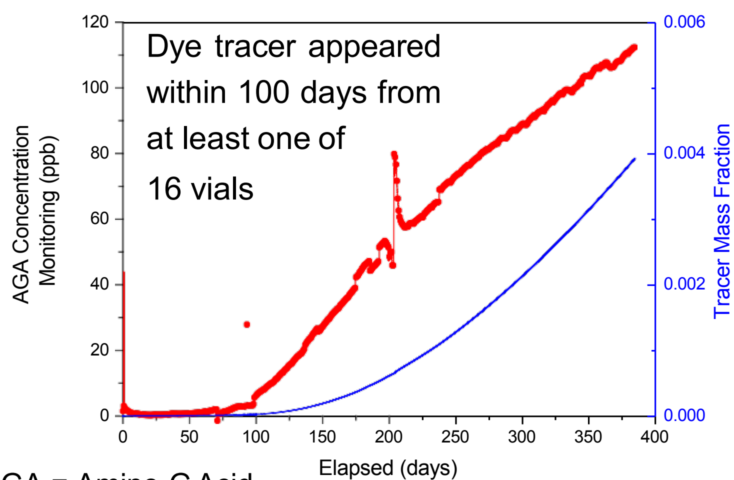
Tracers in glass vials: Ni doped synthetic montmorillonite, AGA and RN:Ca-45, Se-75, Tc-99, Cs-137, U-233, Am-241, Pu-242, Np-237

16 Vials

First ~400 days of monitoring in near-field boreholes

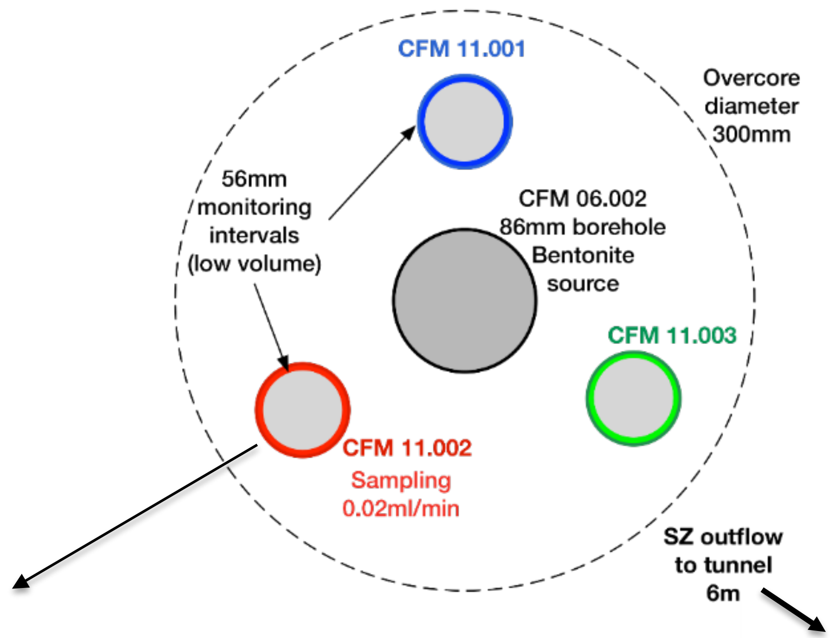
Bentonite saturated and swelled very quickly





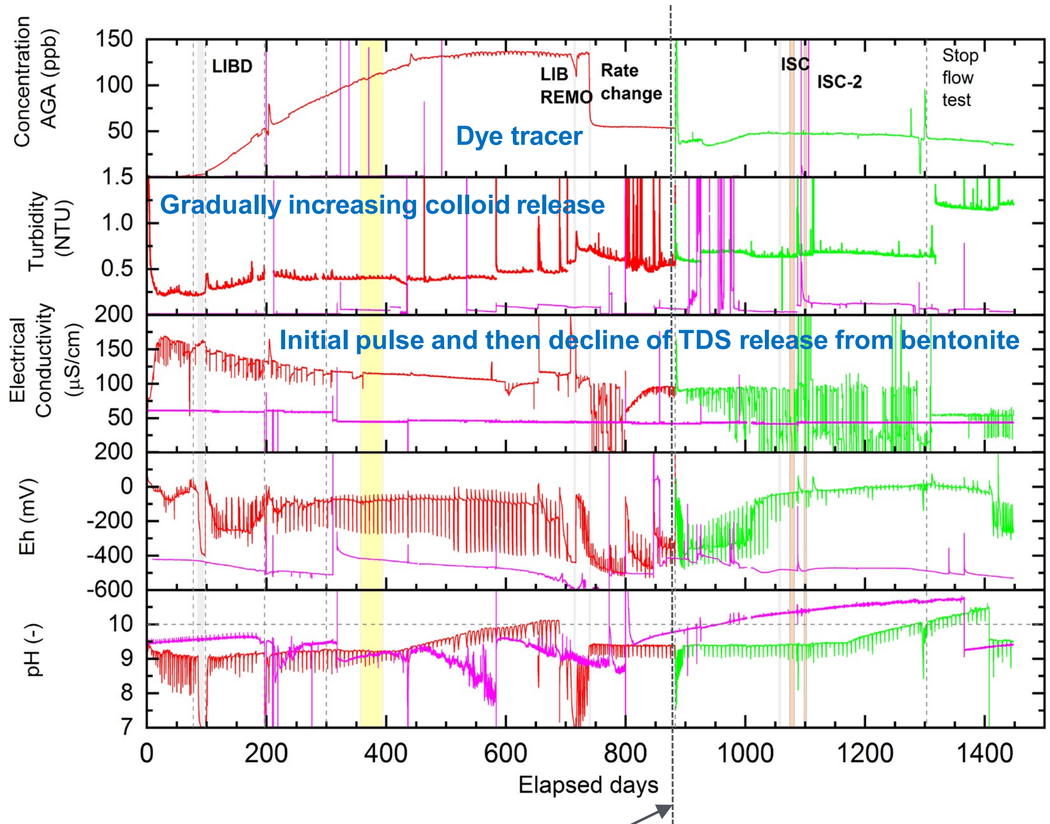
AGA = Amino-G Acid

Overcoring (excavation) initiated in December 2018

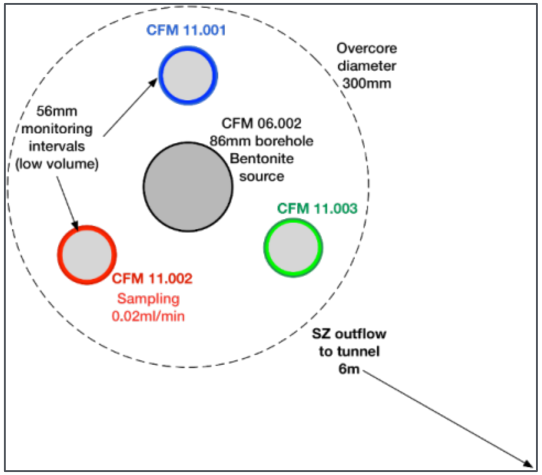


Longer-Term (~4-yr) Results

- Only conservative tracer and minor colloid breakthroughs in monitoring holes (6-7 cm away)
- However, very small concentrations (ppq) of ^{99}Tc are being detected by AMS in monitoring hole
- Almost imperceptible concentrations of conservative tracer and colloids at tunnel wall (~6 m away)
- No actinides detected anywhere



Switch from red to green monitoring hole (882 days)



More detailed information expected from overcoring and post-mortem

Summary of Knowledge Gained from CFM Participation

- Insights have been gained as to how to obtain defensible answers to predict radionuclide transport in fractured granites. However, site specific studies still need to be performed to to gain confidence in the prediction
- The CFT ladder should be applied to evaluate the potential for enhanced transport with colloids, but most indications are that only very small fractions of strongly-sorbing radionuclides will be capable of CFT over repository time and distance scales
- CFT requires very slow desorption from colloids AND very slow filtration of the RN-bearing colloids (relative to time scales of interest)
- Interrogating such slow processes is a challenge, especially if they are associated with a very small fraction of colloids or very small fraction of sorption sites on colloids (or both)
- Intuitively, one might expect that stable colloids generated from waste-form degradation that have radionuclides incorporated into their structure (as opposed to a sorption association) might pose the biggest risk

Summary of Knowledge Gained from CFM Participation (2)

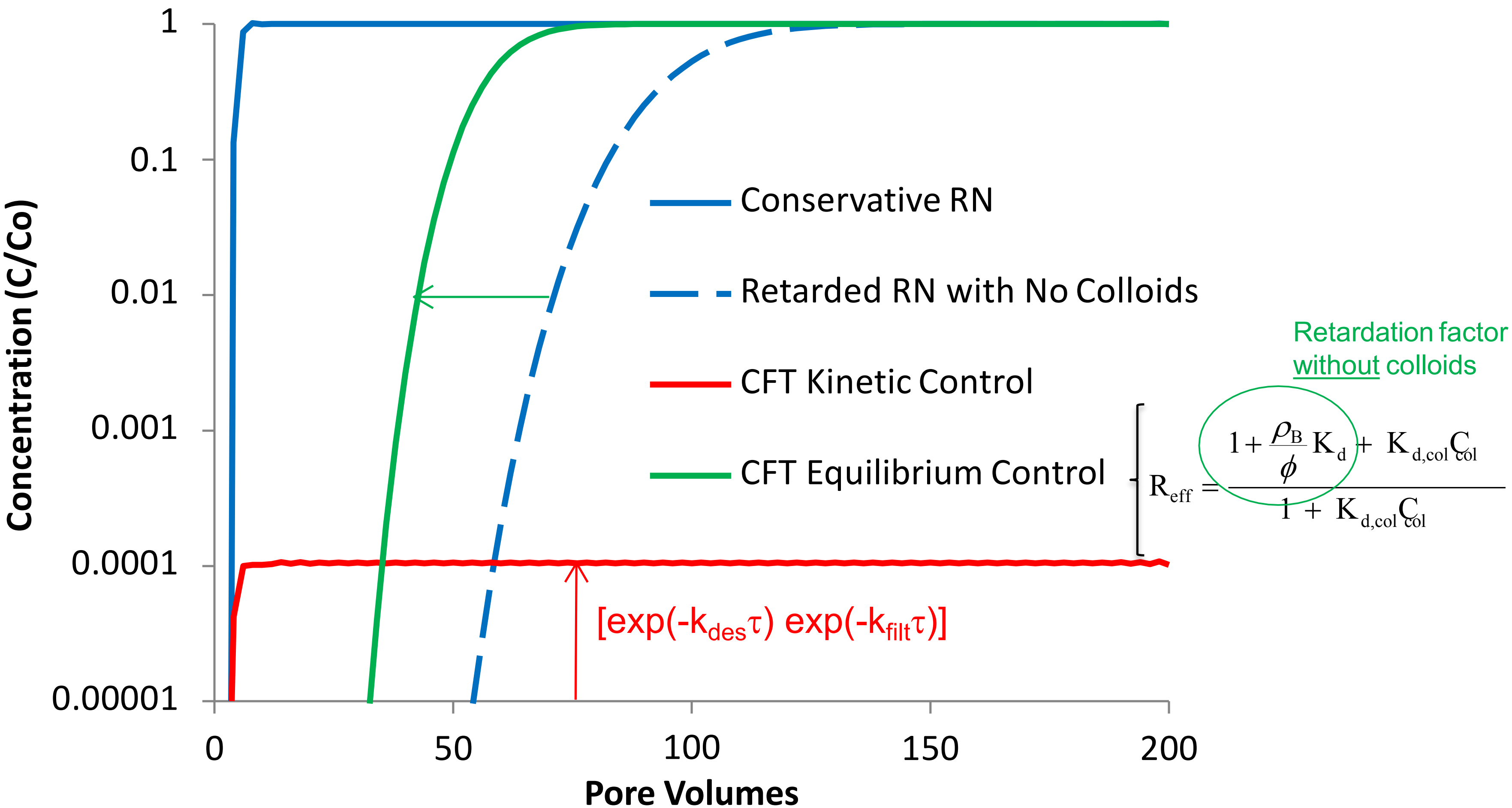
- Experiments have informed generic modeling approach, including GDSA
- Experiments have provided insights into how experimental designs can be tailored and improved to address site-specific and scenario-specific issues
- Different host rocks, EBS vs. natural system, and DPC concept can all, in principle, be addressed via different parameterizations of the generic model, with the understanding that parameterizations must be developed through site- and scenario-specific experimental testing

Refer also to:

Colloid-Facilitated Radionuclide Transport: Current State of Knowledge from a Nuclear Waste Repository Risk Assessment Perspective, *FCRD-UFD-2016-000446*, August 2016.

Mathematical Basis and Test Cases for Colloid-Facilitated Radionuclide Transport Modeling in GDSA-PFLOTRAN, *SFWD-SFWST-2017-000117*, August 2017.

Graphical Depiction of GDSA Approach



For complete model description, refer to:

Mathematical Basis and Test Cases for Colloid-Facilitated Radionuclide Transport Modeling in GDSA-PFLOTRAN, *SFWD-SFWST-2017-000117*, August 2017.

Questions?

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