Sr-90 Immobilization by Infiltration of a Ca-Citrate-PO_4 Solution into the Hanford 100N Area Vadose Zone

Jim Szecsody, John Fruchter, Mark Rockhold, Bob Moore*
Mark Williams, Mart Oostrom, Carolyn Burns, Vince Vermeul

PNNL-SA-61818

Pacific Northwest National Laboratories
*Sandia National Laboratories

Pacific Northwest
NATIONAL LABORATORY

Outline

Background

- Sr-90 contamination in the Hanford 100N area
- EM-22 infiltration project tasks
- financial, status

Proving concepts at small scale

- apatite formation in vadose zone
- Sr-90 incorporation

Infiltration strategy

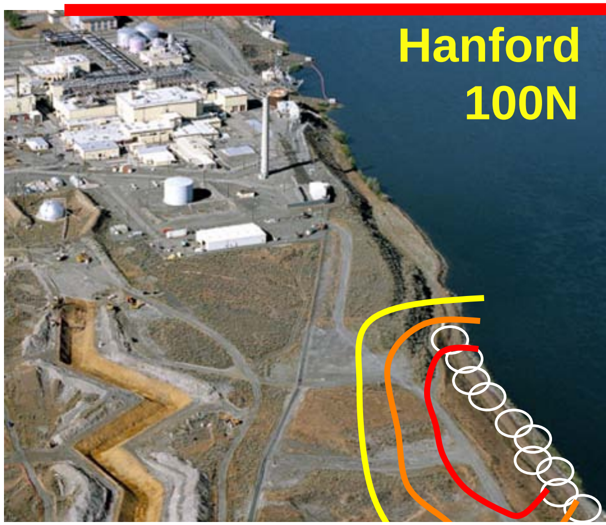
- infiltrate to achieve precipitate at specific depth, in low-K layers
- advection using foam

Upscaling

- large lab scale infiltration (12-ft 1-D, 8-ft 2-D systems)
- microbial transport (30 ft)
- preliminary field characterization, FY09 plan

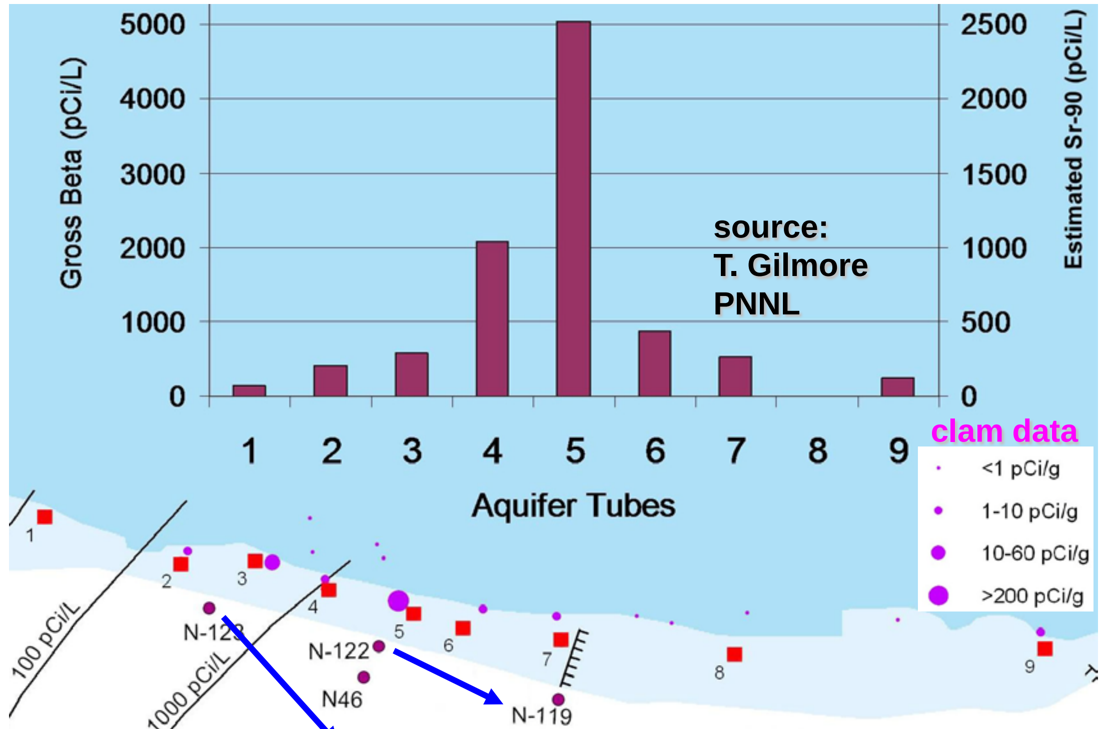
Summary

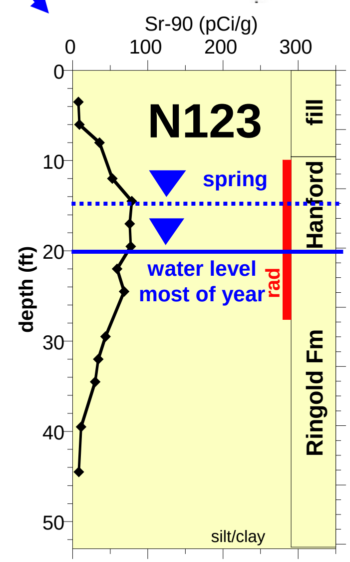
^{90}Sr Contamination Spread

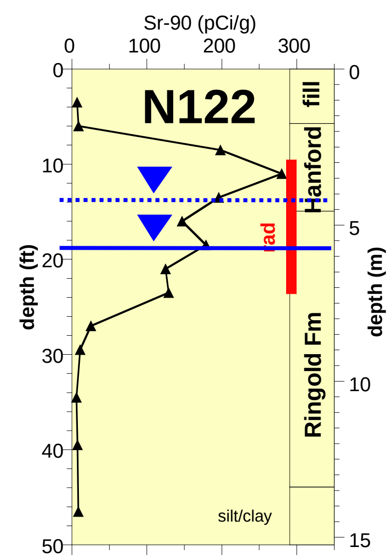


- **Sr-90 in aquifer tubes, clams, and wells** **show high conc. location**
- **Sr-90 in zone of water table fluctuation** **and shallow aquifer (8 - 35' depth)**
- **Sr-90 profile across two formations:**

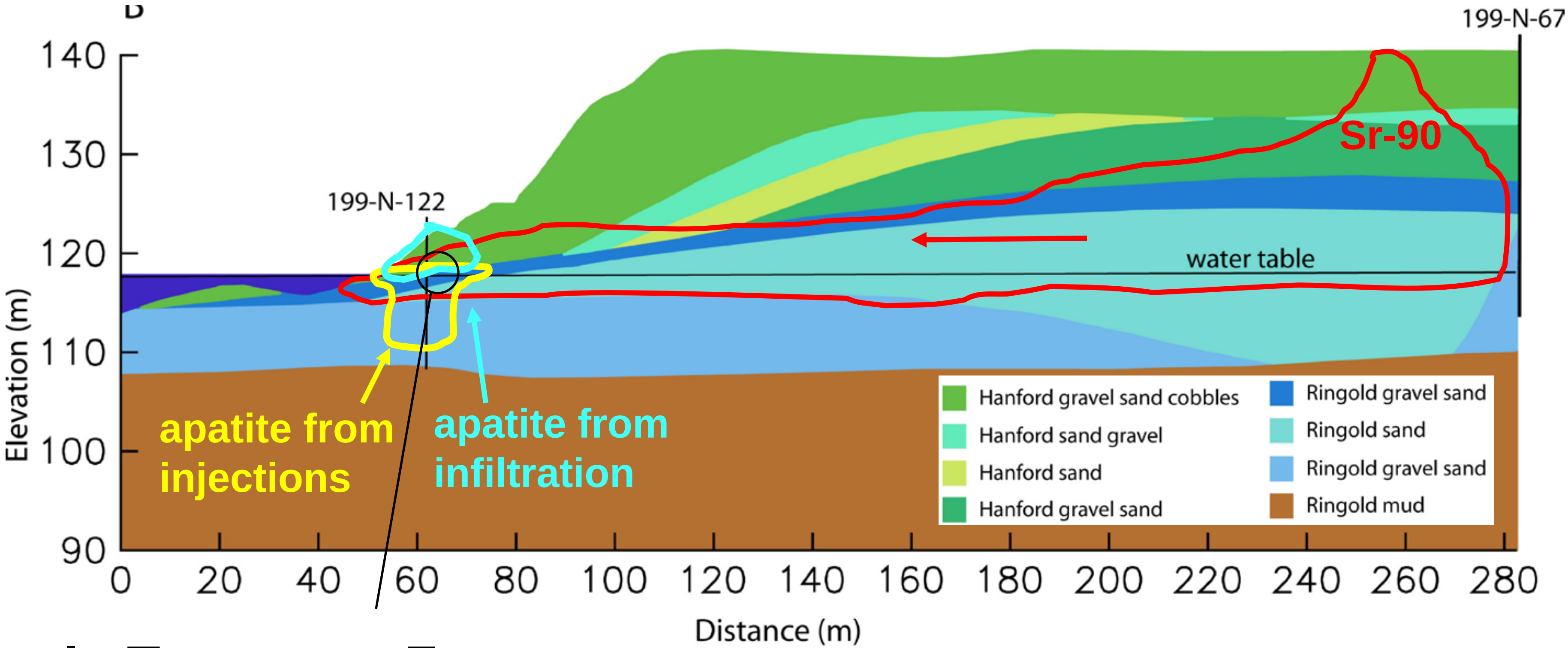
  Hanford Fm. (shallow, high flow)
  Ringold Fm. (deep, 3x low flow)
  which requires differing treatment



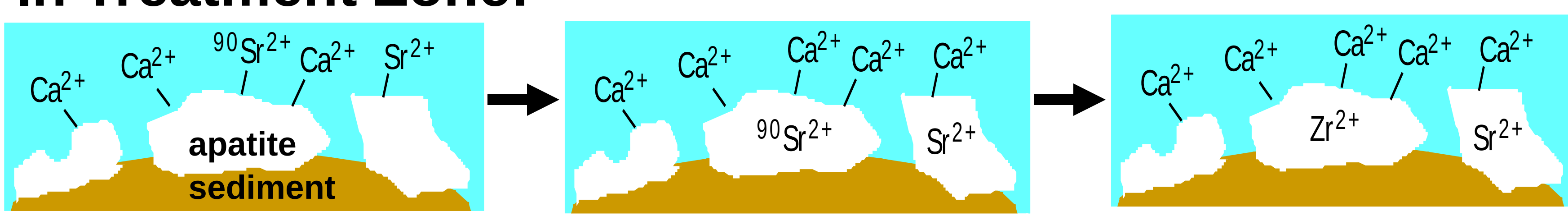




Conceptual Approach



In Treatment Zone:



^{90}Sr adsorbs (minutes)

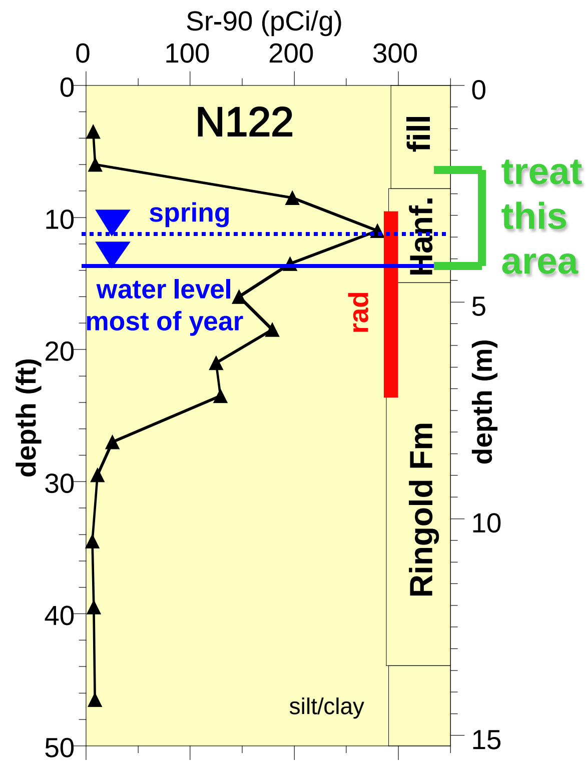
^{90}Sr incorporates into apatite (months, yrs)

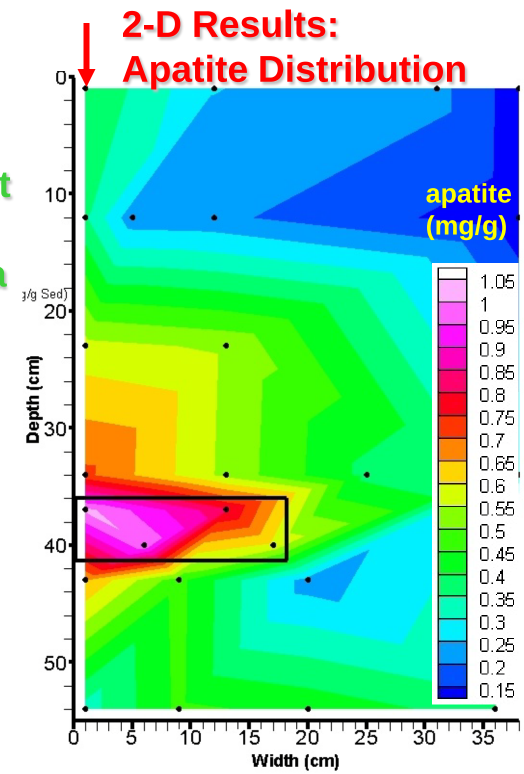
decay (10s, 100s yrs)

$^{90}Sr \xrightarrow[29.1yr]{t_{1/2}} {}^{90}Y \xrightarrow[60hr]{} {}^{90}Zr$

Objectives

- Immobilize Sr-90 by incorporation into hydroxyapatite
  $Ca_{10}(PO_4)_6(OH)_2 \; \text{-->} \; Ca_{(10-x)}Sr_x(PO_4)_6(OH)_2$
- Emplace apatite by Ca-citrate-PO_4 solution infiltration into the vadose zone (30-70% of Sr-90 mass)
- Develop infiltration strategy for the Ca-citrate-PO_4 solution to result in apatite precipitation at:
  - specific depth
  - in low-K zones
- test with 1-D/2-D experiments with modeling



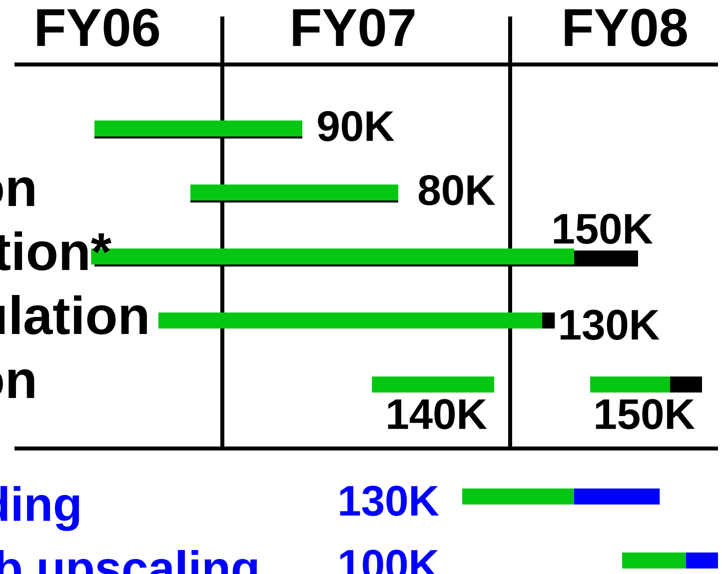


Solution Infiltration and Sr-90 Immobilization Tasks and Schedule

batch → 1-D → 2-D (1.5m) → field infiltration design → 2-D (4 m) → field test

Task:

1. Apatite Fm. — completed
2. 1-D Infiltration — completed
3. Sr Incorporation* — with SNL
4. 1-D/2-D Simulation — completed
5. 2-D Infiltration — in progress



Task 5 EMSL funding — 130K

Fluor funding - lab upscaling — 100K

collaboration with Env. Biomarkers Initiative Project (G. Bonheyo)

Fluor funding - field infiltration — 50K

Task Leads:

1. J.Szecsody, C. Burns
2. J. Szecsody, M. Oostrom
3. C. Burns, B. Moore* (SNL)
4. M. Rockhold
5. M. Oostrom, J. Szecsody

Szecsody, J, C. Burns, R. Moore, J. Fruchter, V. Vermeul, D. Girvin, J. Phillips, M. Williams, 2007, Hanford 100N Area Apatite Emplacement: Laboratory Results of Ca-Citrate-PO4 Solution Inject and Sr-90 Immobilization in 100N Sediments, PNNL report. PNNL-16891.

Szecsody, J., J. Fruchter, C. Burns, M. Rockhold, M. Oostrom, M. Williams, V. Vermeul, and R. Moore, 2008, Sr-90 Immobilization by Infiltration of a Ca-Citrate-PO4 Solution into the Hanford 100N Area Vadose Zone. Waste Management 2008 Conference, Phoenix, AZ

J. Szecsody, V. Vermeul, M. Williams, J. Fruchter, B. Moore, C. Burns, 2008, Sequential Injections of a Ca-Citrate-PO4 Solution into Aquifer Sediment for Sr-90 Immobilization in preparation

R. Moore, J. Szecsody, C. Burns, V. Vermeul, J. Fruchter, M. Williams, 2008, Optimization of a Ca-Citrate-PO4 Solution Used for Sr-90 Immobilization in a Groundwater System in preparation

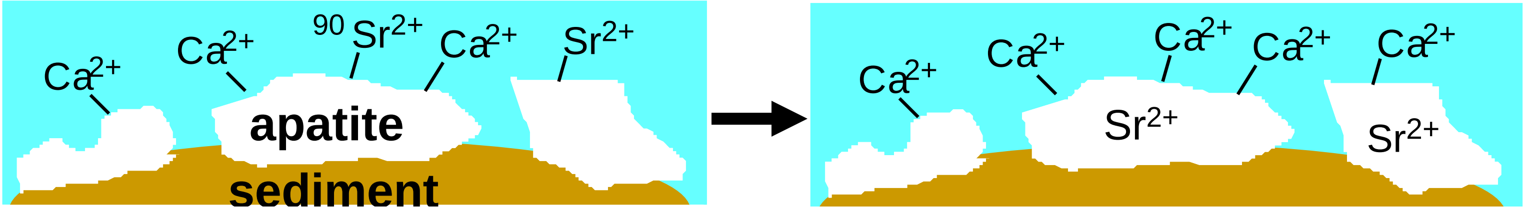
Earned Value Report

Apatite Infiltration	June 08	FY08
BCWS	$44,956	$713,131
BCWP	$11,250	$683,862
ACWP	$8,700	$680,699
SV	-$33,706	-$29,269 behind schedule*
CV	$2,550	$3,163

* additional Fluor-funded 2-D tasks (FY08), delaying 29K EM22 funding to FY09

Apatite Sequestration - Background

- Calcium phosphate minerals (apatite; $Ca_5 (PO_4)_3X$) are stable and very insoluble [X = OH-, Cl-, F-]
- Apatite easily incorporates a number of cationic contaminants [Pb, Sr, Cd] in place of the Ca

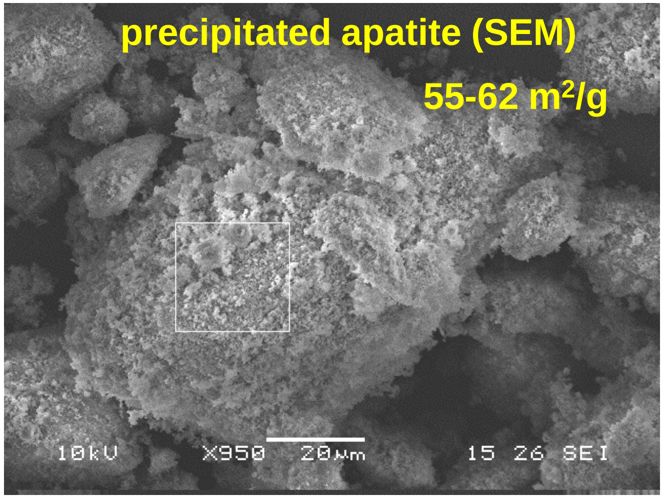


- Some natural apatites are as high as 11% Sr
- Solid apatite has been used to absorb these contaminants in a variety of applications

Apatite Formation in Sediment

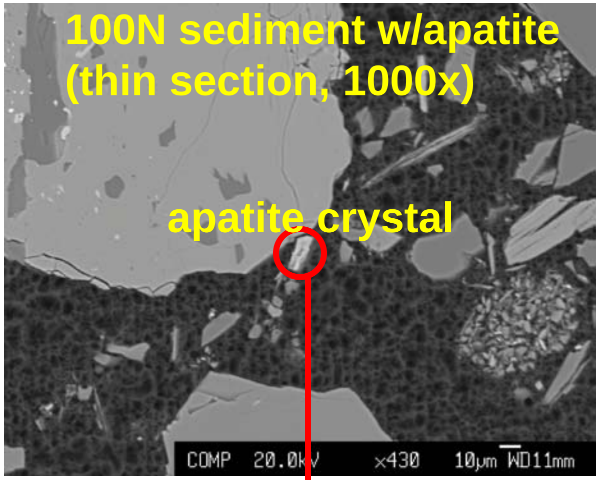
- **Infiltration solution:**
  Ca-citrate complex, Na_2HPO_4 (pH 7.7)
  stable for 24-36 h without precipitation

$Ca^{2+} + PO_4^{3-}$
ppt within seconds

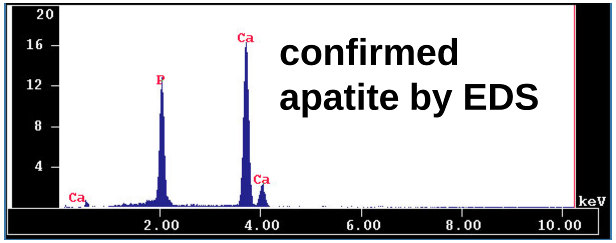


- **Solution contact with sediment:**
  citrate biodegrades (half-life ~50 - 150 h)
  releasing Ca^{2+}, and at this pH apatite forms

$$Ca^{+2} + HPO_4^{-2} \longrightarrow Ca_{10}(PO_4)_6(OH)_2$$



- **Precipitate formed confirmed hydroxyapatite**
  by SEM/EDS, XRD
  (amorphous at 1 week, crystalline in 3-4 weeks)

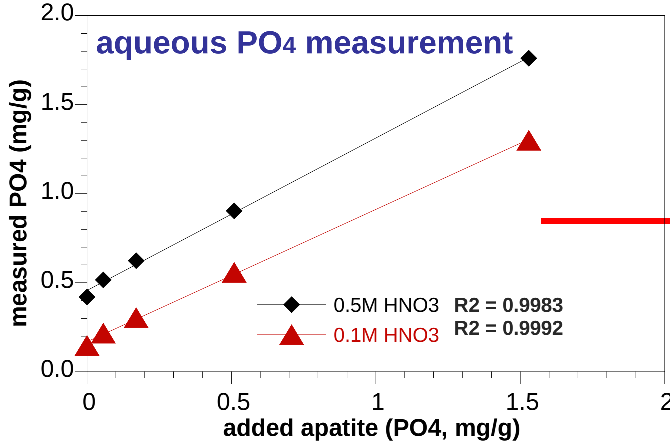


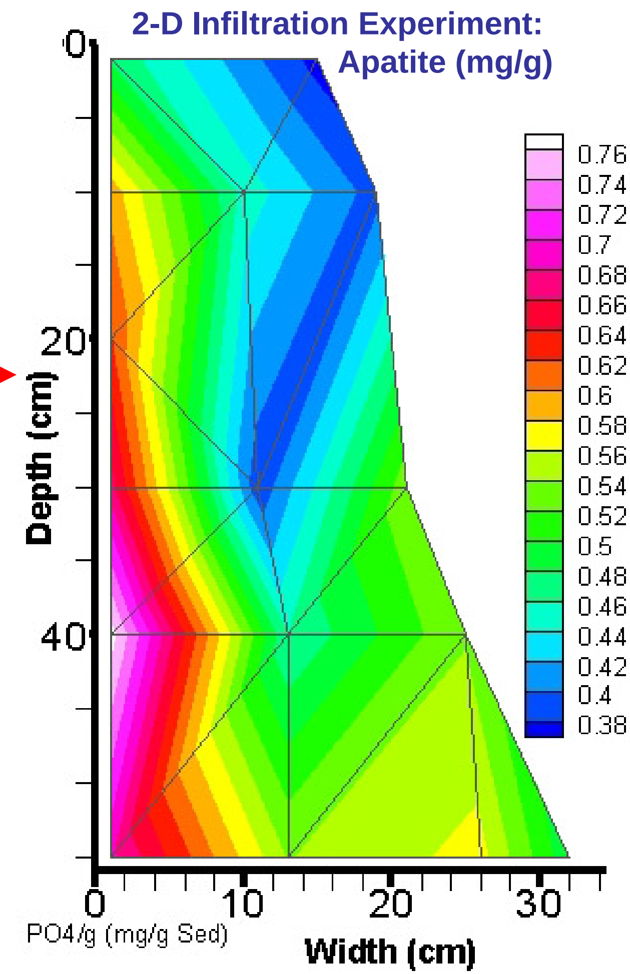
Task 1: Measuring Apatite Precipitate in Sediment

A small amount of apatite precipitate is produced (0.1 - 3 mg/g), which can be quantified by:

1. electron microprobe w/EDS detector

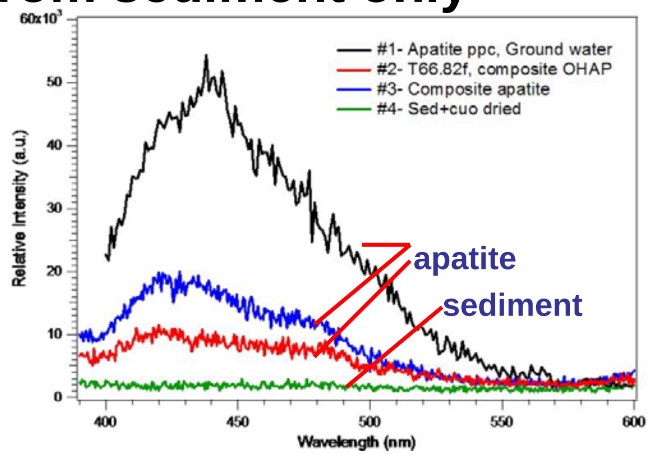
2. acid dissolution/ PO_4 measurement





3. fluorescence of various apatites, no fluorescence from sediment only

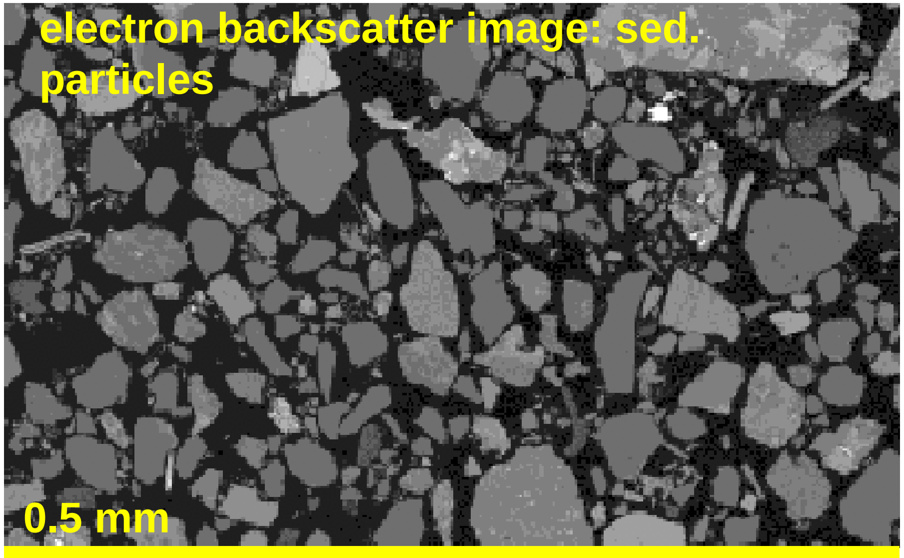
Fluorescence from F- and CO_3^{-2} substitution in apatite

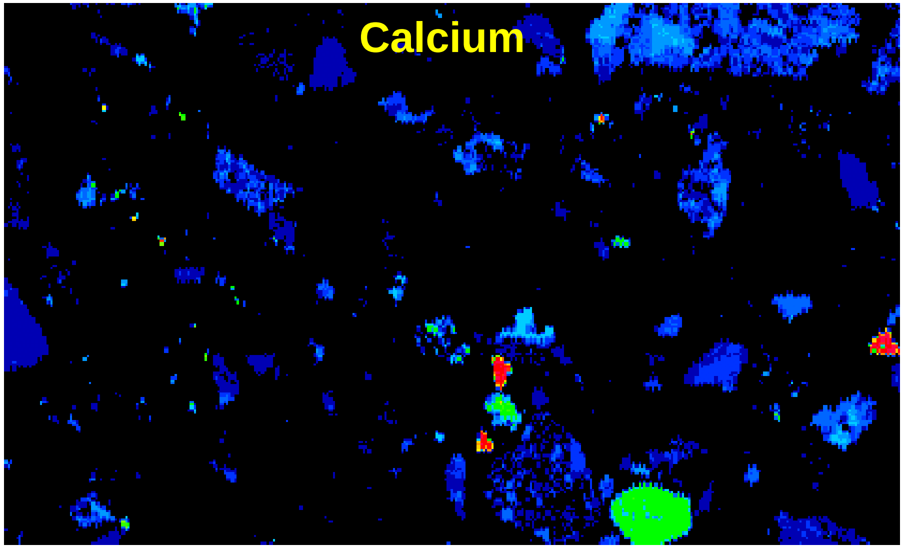


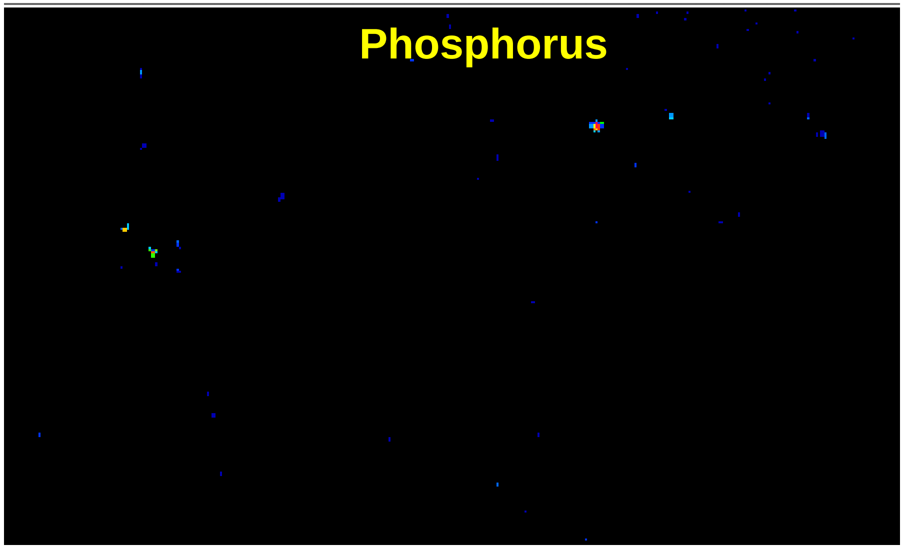
Task 1. Identify Small Mass of Apatite in Sediment

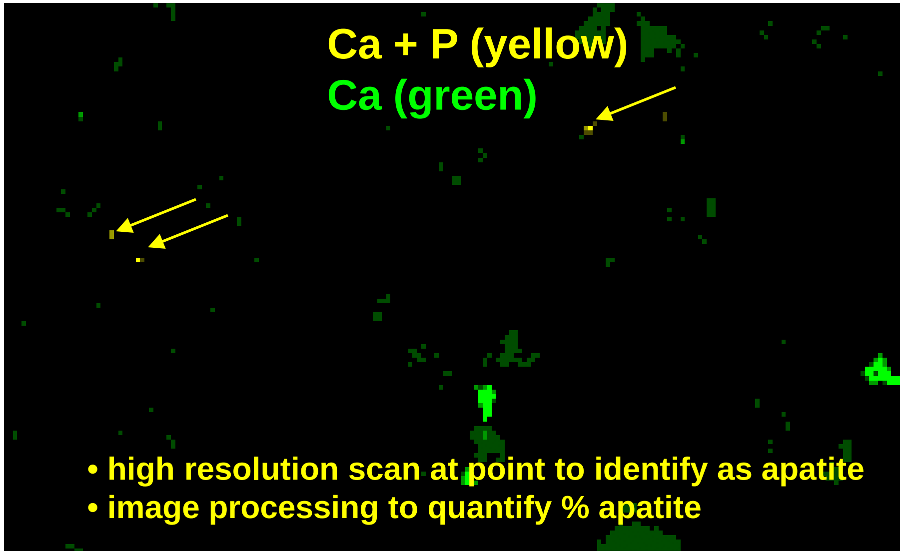
sample: 0.05 mg apatite/g sediment (equivalent to injection #1, #2: 2.4 mM PO4)
data: electron microprobe, elemental spectrum detectors

microprobe analysis: J. McKinley





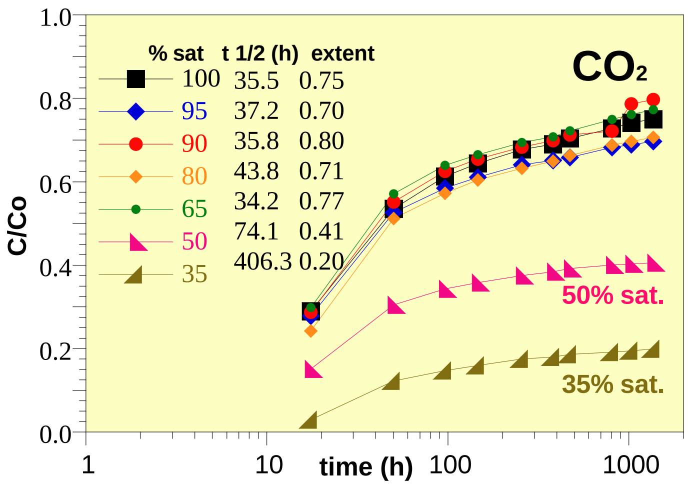




Task 1: Citrate Degradation

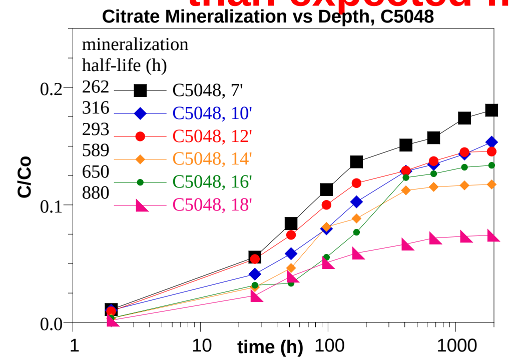
occurs at low water sat.

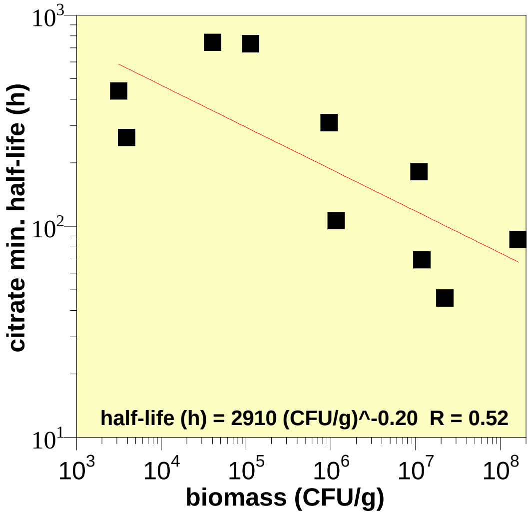
citrate aerobically degrades to CO_2; slower at low water sat.

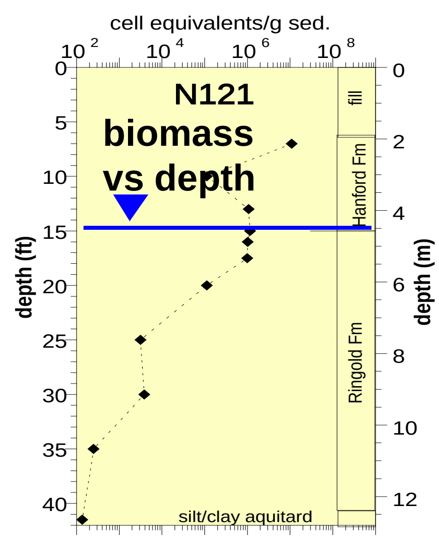


rate more uniform than expected

mineralization rate is more uniform than expected from biomass





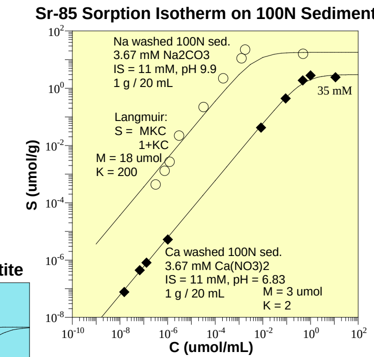


- **Questions:**
  - in situ vs infiltrating microbes?
  - microbial transport?

Sr-90 Mobility in Sediment/Apatite

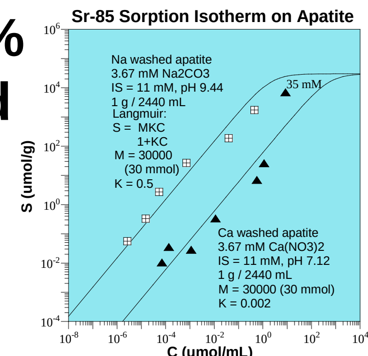
- Sr sorbs to sediment: 1% aqueous, 99% sorbed by ion exchange - easily removed

Sr-90/sediment: $K_d = 24.8 \pm 0.4\ cm^3/g$ →

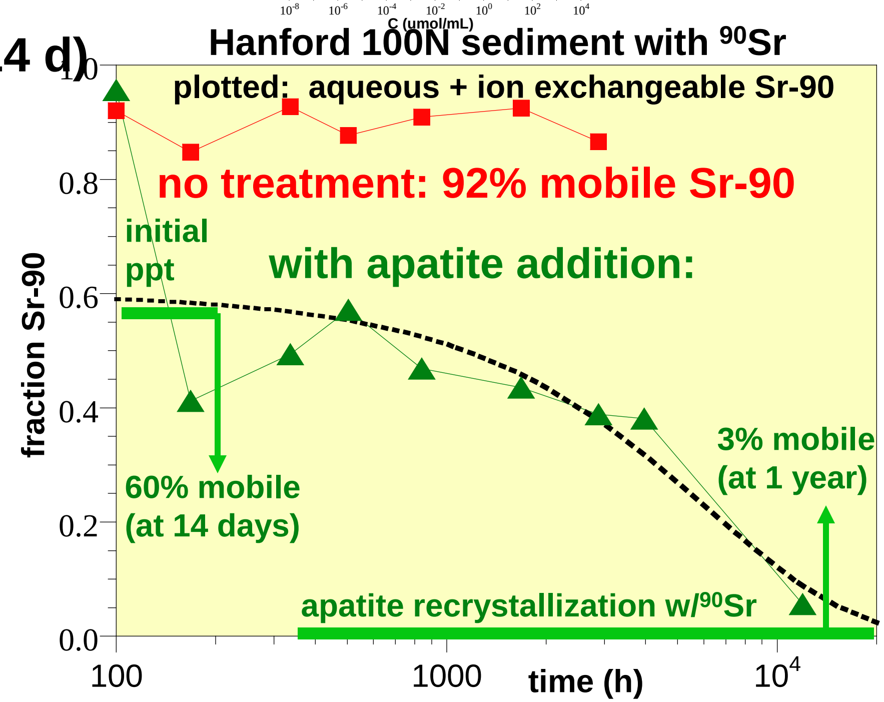


- Sr sorbs to apatite: 0.02% aqueous, 99.98% sorbed by ion exchange - easily removed

Sr-90/apatite: $K_d = 1370 \pm 429\ cm^3/g$ →



- Sr is slowly incorporated into apatite:
  - -during initial apatite precipitation (< 14 d) (source zone treatment)
  - -during recrystallization (72-180 d $t_{1/2}$) (permeable reactive barrier)



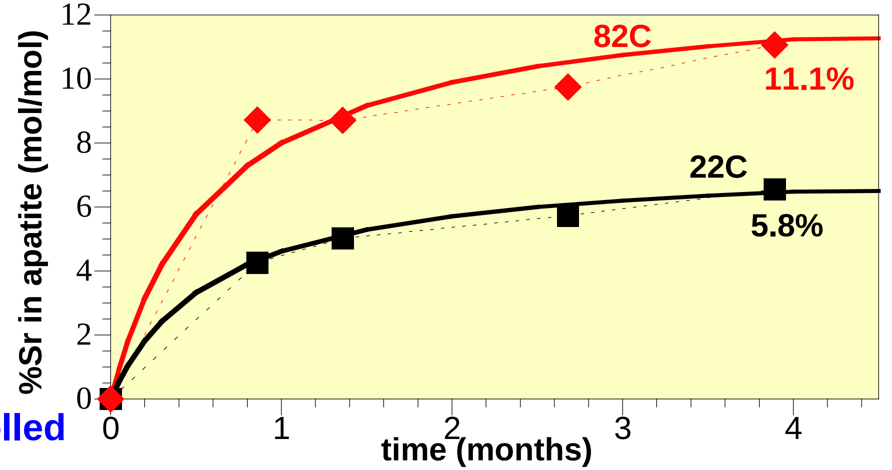
- Need to immobilize Sr-90 for 300 yrs

Ca/Sr-apatite is extremely insoluble

[$\log K_{sp(Ca\text{-}apatite)} = 10^{-44}$, $\log K_{sp(Sr\text{-}apatite)} = 10^{-51}$]

Sr-90 Incorporation into Apatite

Sr-90 is incorporated into apatite:
- a) during initial precipitation (days)
- b) by slow exchange within solid phase (months) →

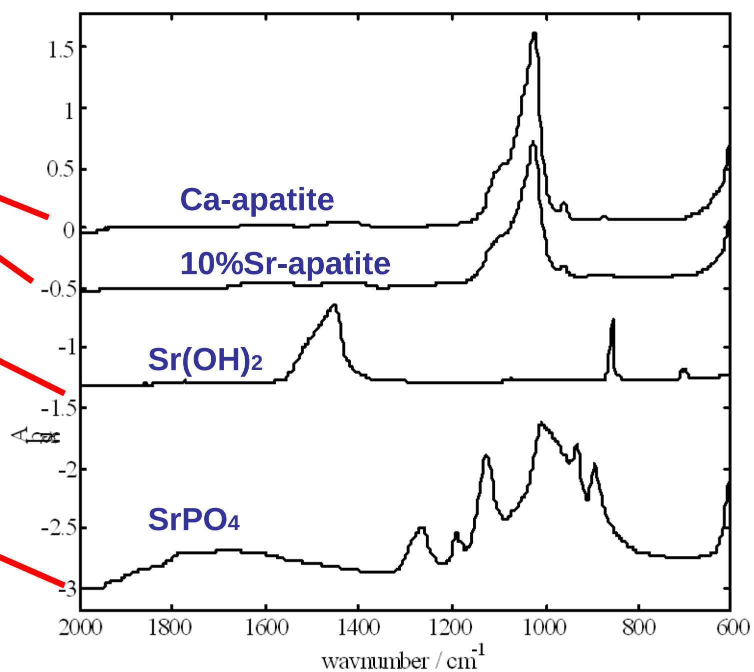


- up to 11% Sr substitution for Ca measured (multiple methods of solid phase extraction)
- activation energy 11.3 kJ/mol; likely diffusion controlled

Apatite crystal structure is not changed by Sr substitution (still hydroxyapatite)

- Ca-apatite has same crystal structure as Sr-apatite (same peaks; process is substitution)
- surface $Sr(OH)_2$ is not precipitating (different peaks from apatite, standard $Sr(OH)_2$ shown)
- mono- and di-Sr-PO_4 not precipitating (different peaks from apatite)

FTIR Scan of Precipitates



Apatite Mass Needed to Sequester Sr-90

Mass Balance, assume:

1. 10% substitution of Sr for Ca in apatite (easily achieved in lab exp)
2. capture all Sr and Sr-90 for 300 years (10 Sr-90 half-lives)

-> need 3.4 mg apatite/g of sediment (i.e. 90 mM PO_4)

Sr-90 Uptake Rate:

- Sr-90 incorporation rate into apatite must be faster than Sr-90 flux rate in aquifer toward the river
  assume: 1 ft/day gw flow, Rf = 125
  15 ft apatite treatment zone radius

Sr groundwater flux rate (ave.) = 1.4×10^{-6} mmol Sr day^{-1} cm^{-2}

Sr incorporation rate (0.34 mg/g) = 8.8×10^{-6} mmol Sr day^{-1} cm^{-2}

(3.4 mg/g) = 8.8×10^{-5} mmol Sr day^{-1} cm^{-2}

-> need 3.4 mg apatite/g of sediment to uptake all Sr-90 for 300 years even in most high-K (10x) zones; 100x flow zones would exceed uptake rate

Simulation of Solution/Sediment Interactions

objective: quantitatively describe reactions and transport (lab, field)

(reactions in code STOMP)

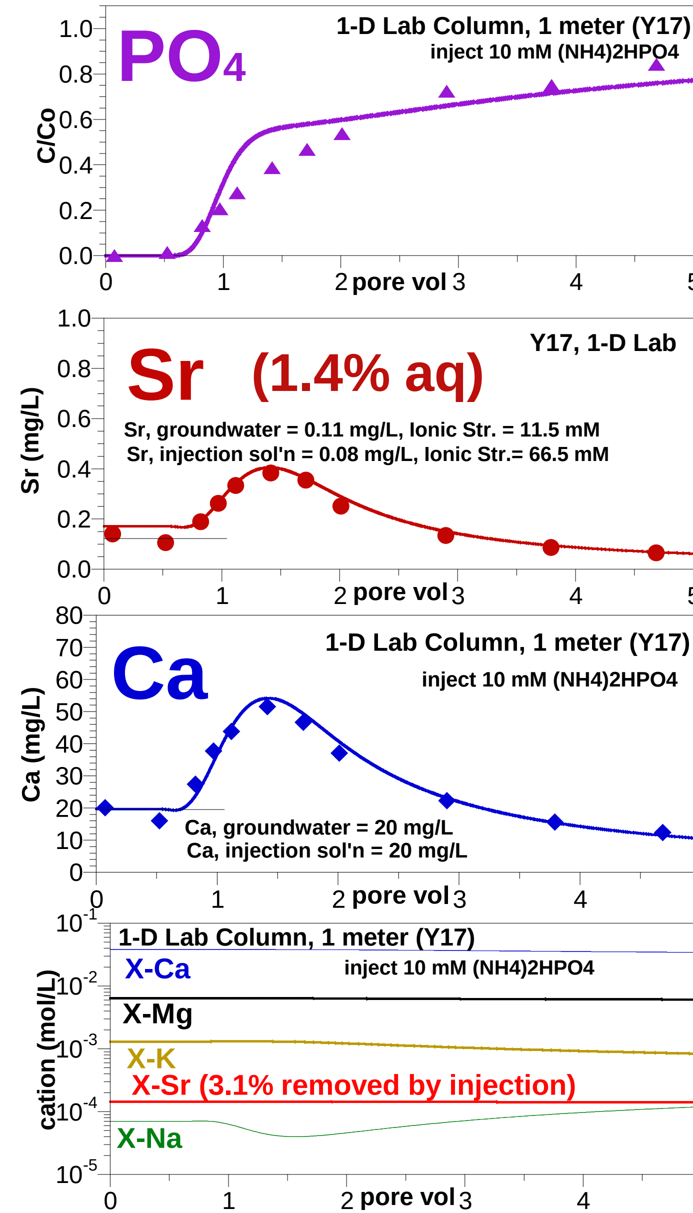
42 reactions, 51 species:

Sr, Ca, Mg, Na, K, NH_4 ion exchange
metal-CO_3 aq. speciation
metal-OH aq. speciation
metal-PO_4 aq. speciation
solids: apatite, $CaCO_3$, $SrCO_3$
citrate biodegradation
(Sr, Ca, Mg, Na)-citrate aq. speciation

physical: 3-D, saturated/unsaturated

simulation results (1-D, saturated):

- ion exchange parameters highly sensitive
- percent Sr or Sr-90 mobilized very small



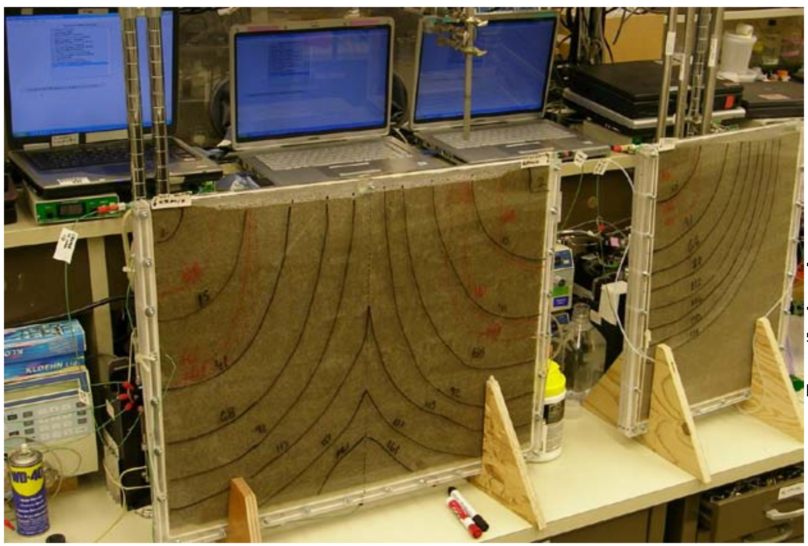
2-D Infiltration to Form Apatite

- **Hypothesis:**

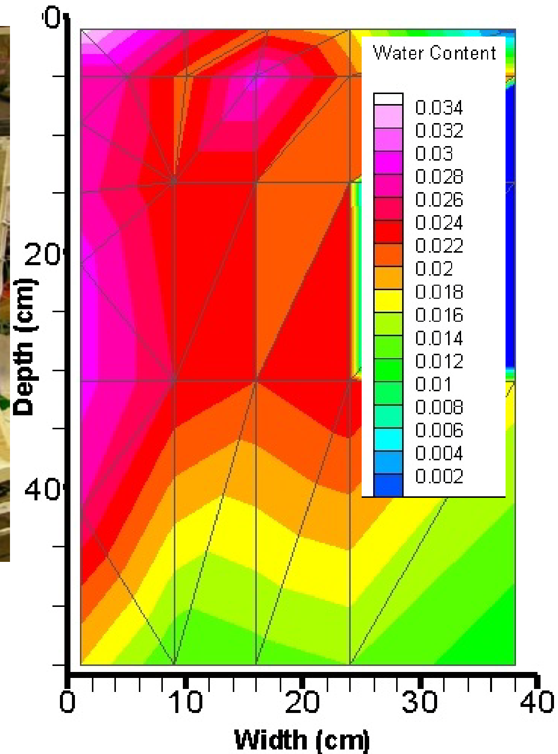
  Chemical/microbial and physical processes control precipitate location:

  - slow infiltration: near surface precipitation only
  - 50-200 h infiltration: should precipitate as front advances (~uniform ppt)
  - rapid infiltration: transport reactants to depth, but little lateral movement

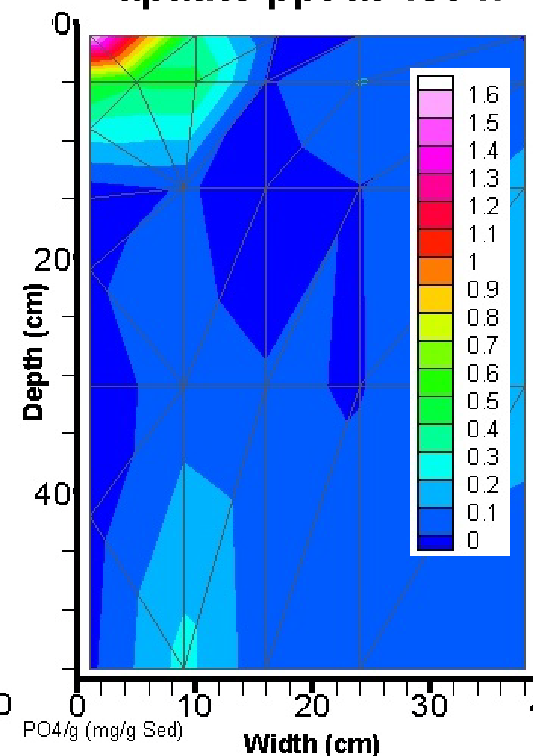
- **2-D lab experiments varying infiltration rate, solution conc., and flow sequence**



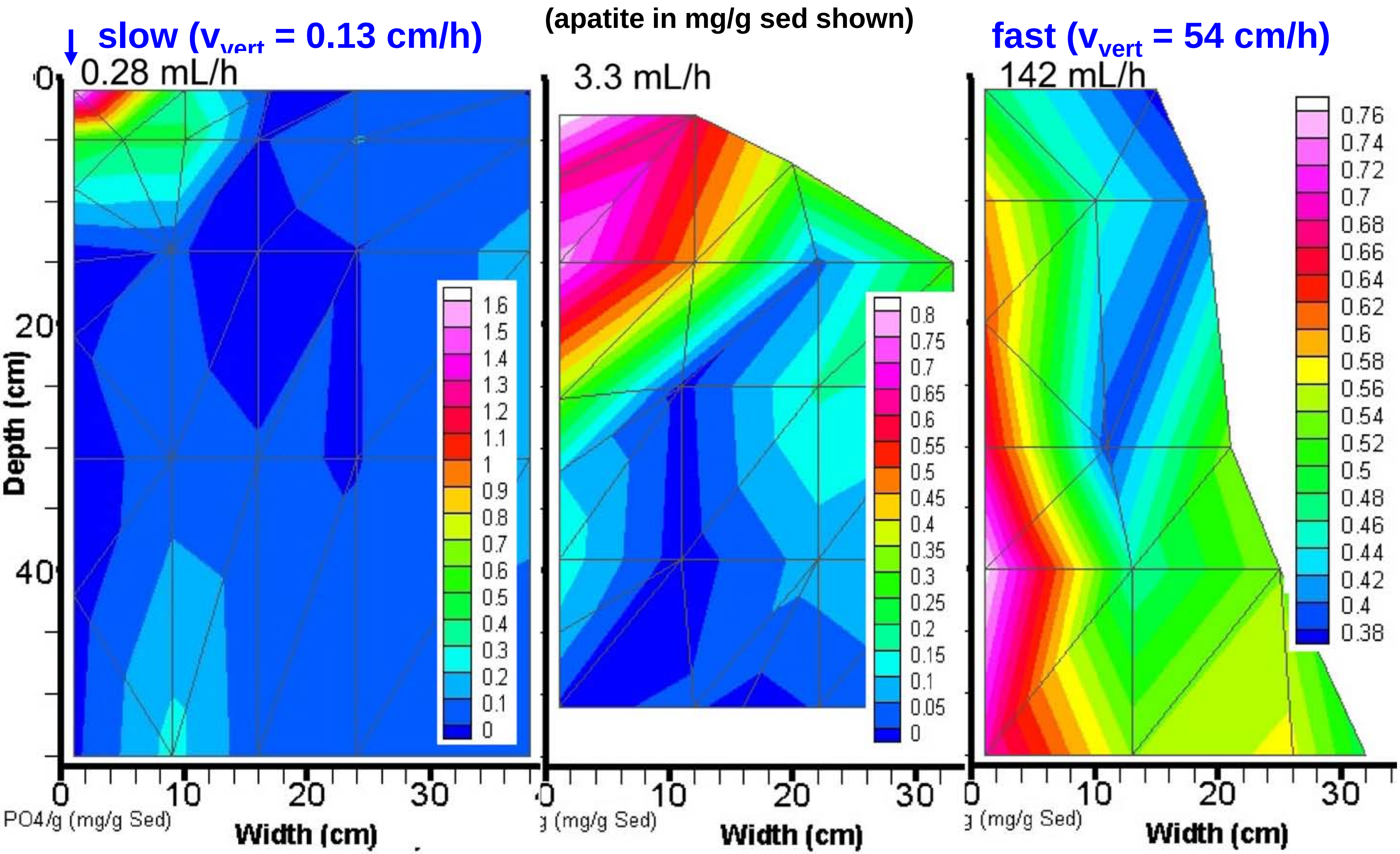
water content at 430 h



apatite ppt at 430 h

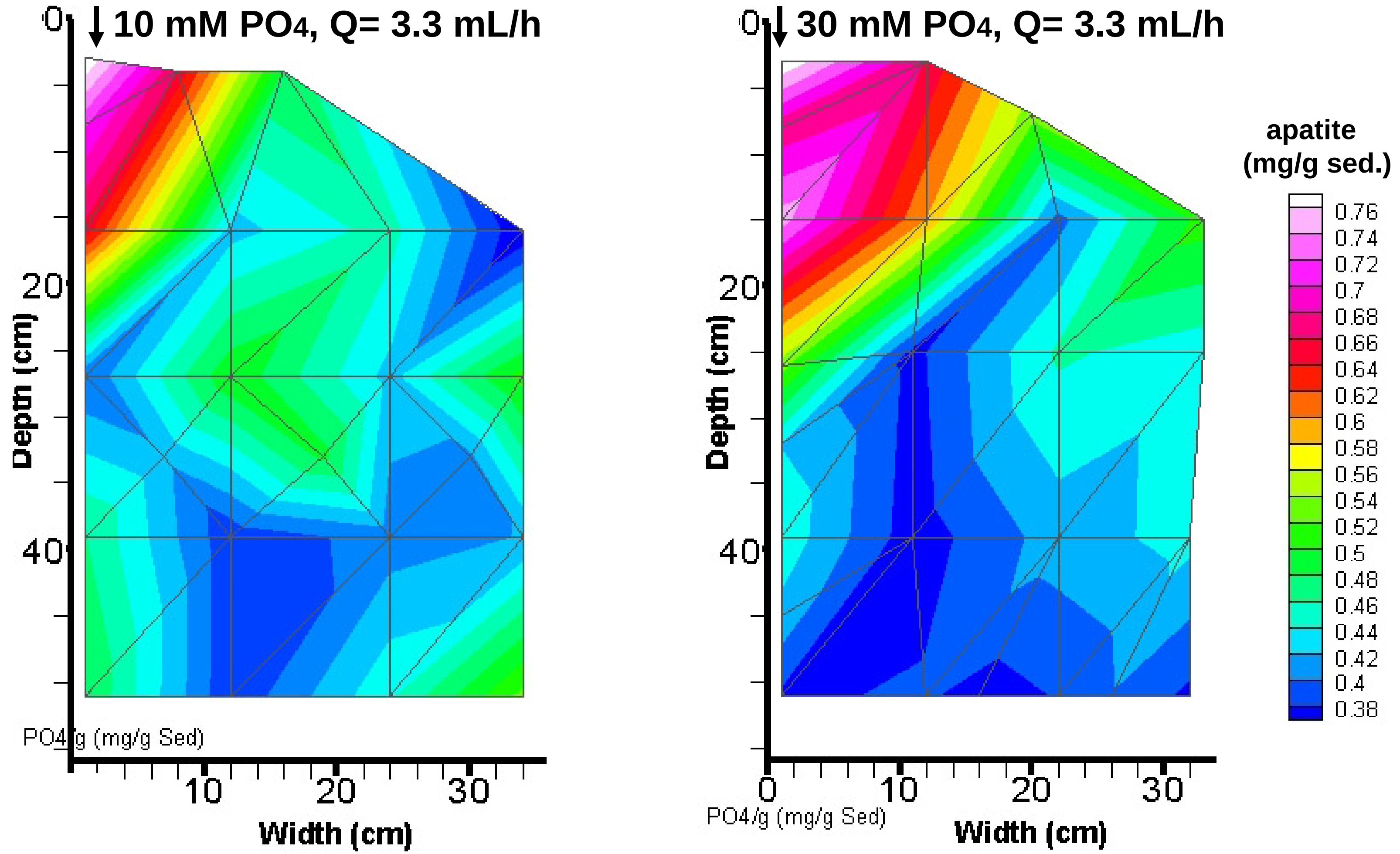


2-D Infiltration: Vary Infiltration Rate



- PO4 transport controlled by adsorption (hours) and citrate biodeg./Ca-PO4 ppt (150 h), so rapid infiltration can override reaction kinetics
- dynamics of vertical/horizontal unsaturated flow define precipitate shape

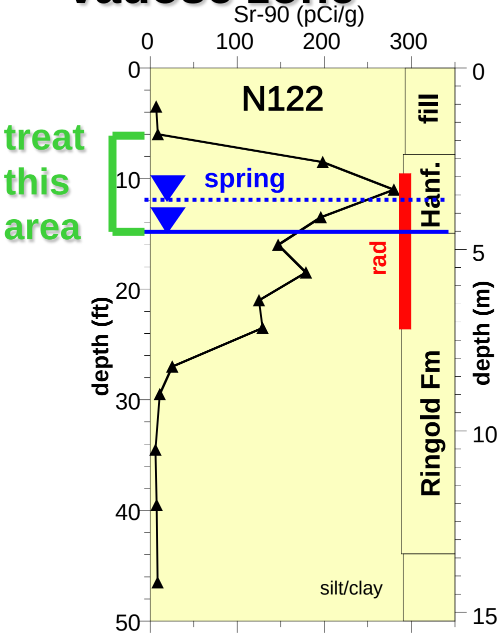
2-D Infiltration: Vary Solution Concentration



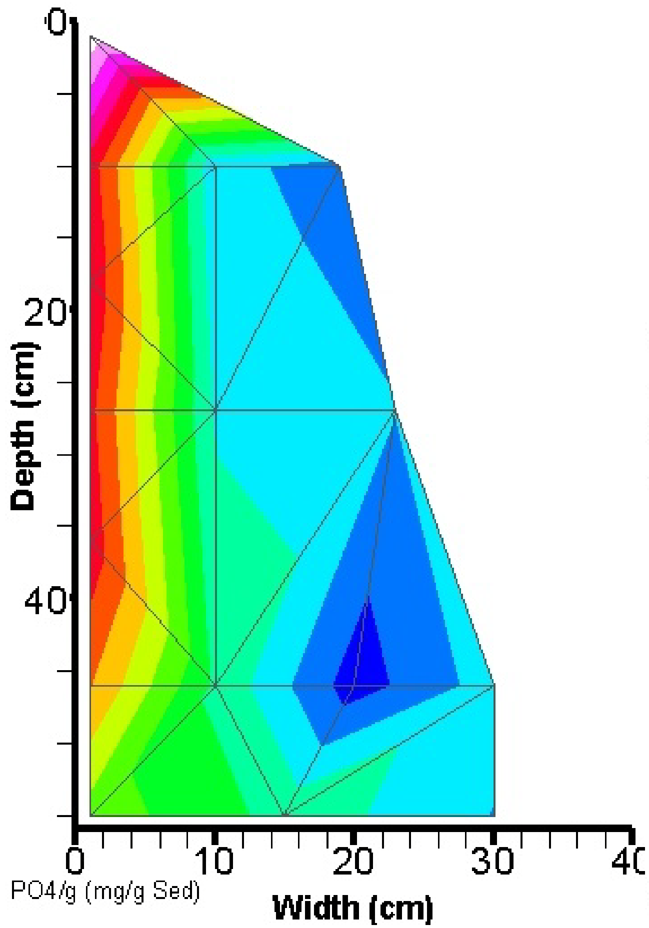
- higher PO_4 conc. results in more area coverage of apatite ppt
  - less PO_4 sorption
  - slower citrate biodegradation

2-D Infiltration: Need Precipitate at Depth

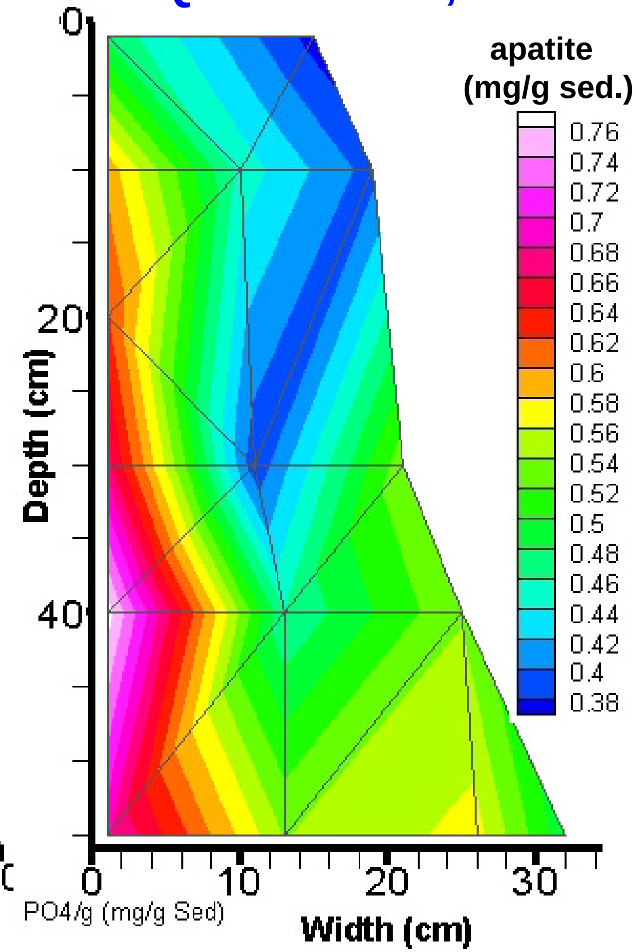
Sr-90 profile: treat lower half of vadose zone



Q= 141 mL/h, 30 mM PO_4



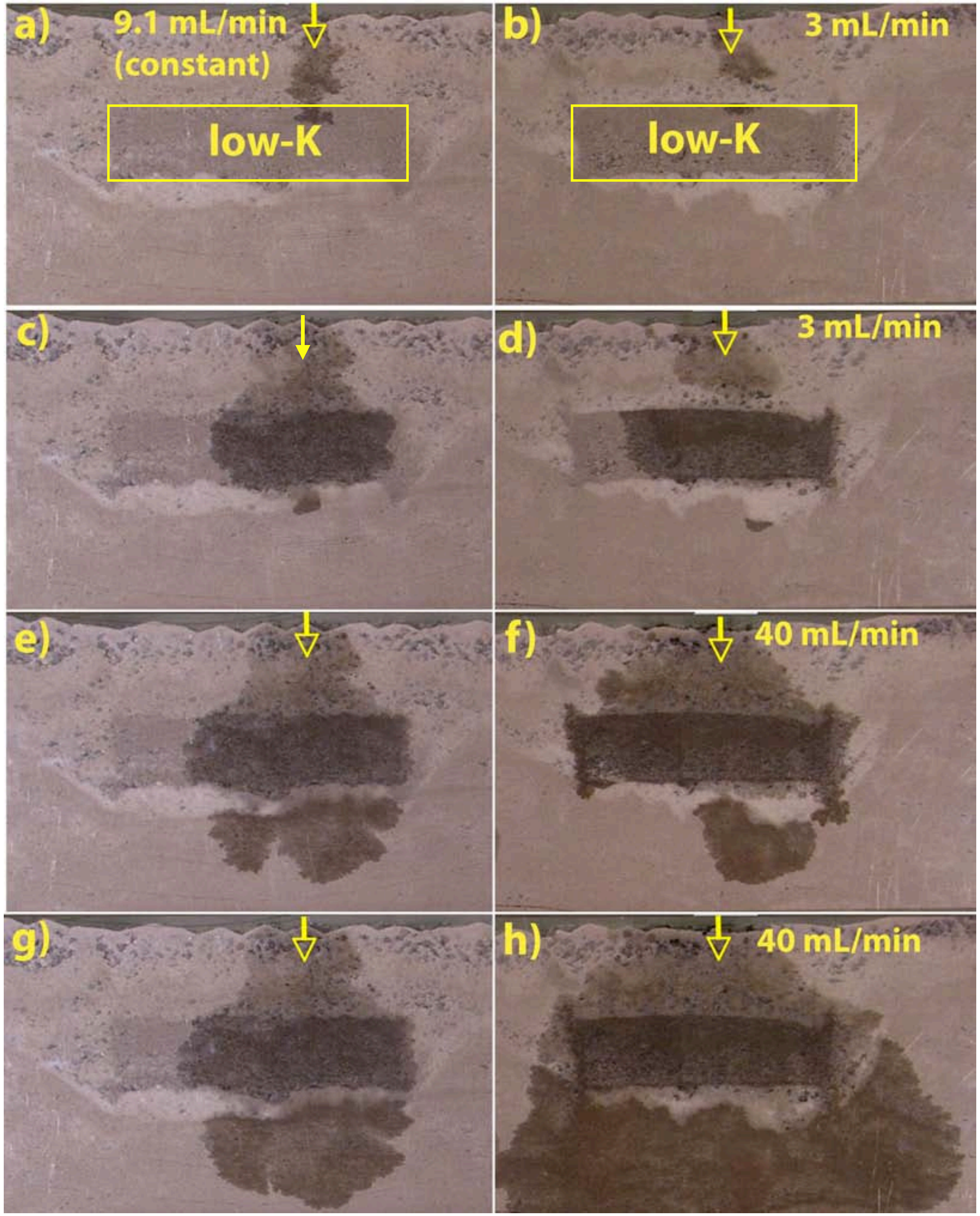
Q= 141 mL/h, 30 mM PO_4
then Q= 141 mL/h, H_2O



- **manipulate infiltration concentration and flow rate to achieve apatite precipitate in desired deeper location**

2-D Infiltration: Need Precipitate in Low-K Zones

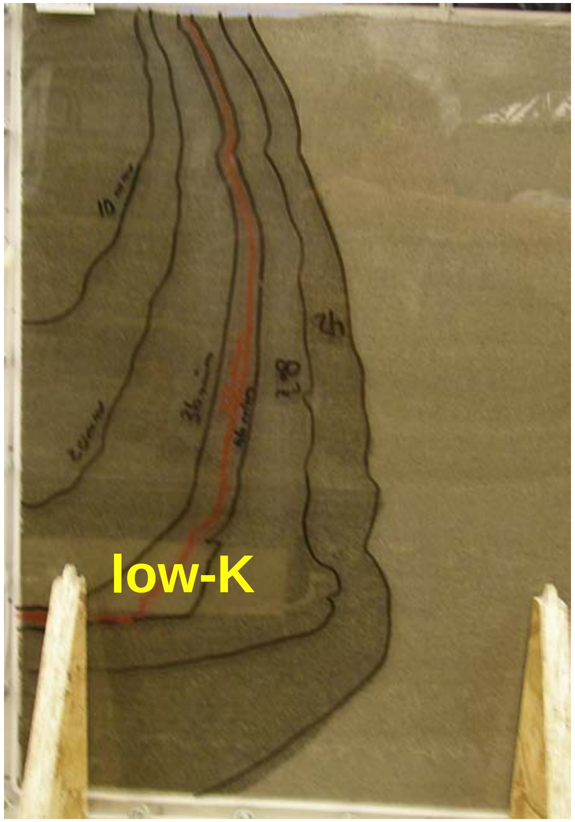
- **Low-K zones likely contain higher Sr-90 concentration**
- **Greater solution coverage in and under low-K zones can be achieved by flow manipulation:**
- **To apply in field, need real-time information of wetting front (borehole and surface geophysics)**

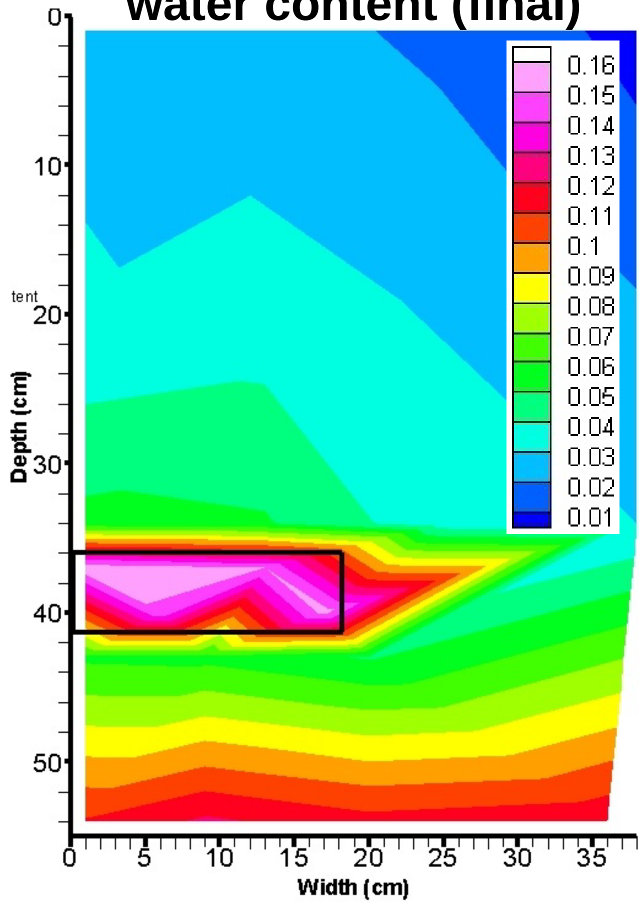


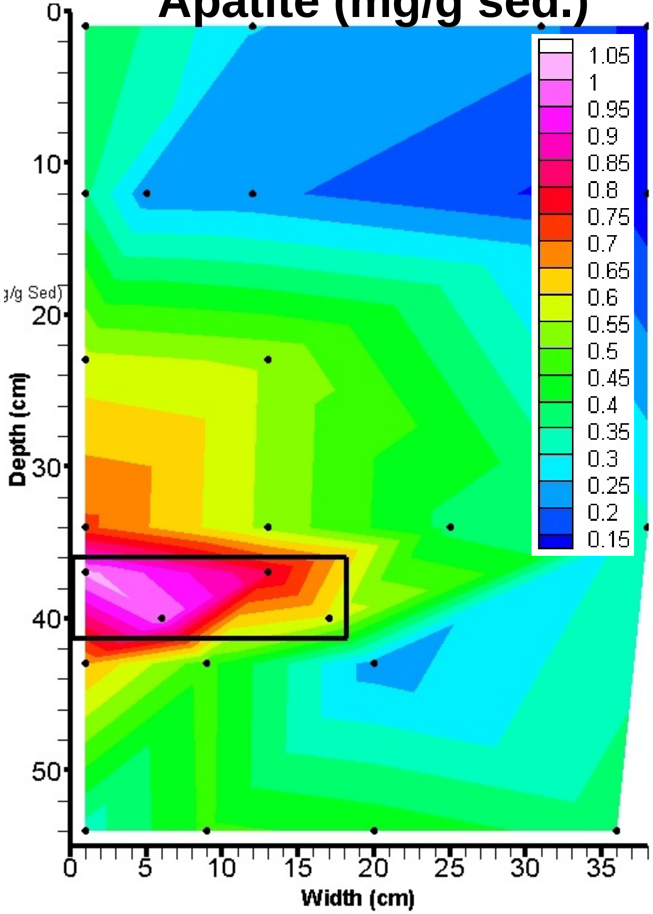
2-D Infiltration with Low-K Zone

- **Hypothesize more Sr-90 in low-K layers/lenses (Kd higher, less flow)**

Q= 141 mL/h, 30 mM PO_4 (1h)
Q= 1.0 mL/h, water (42h)



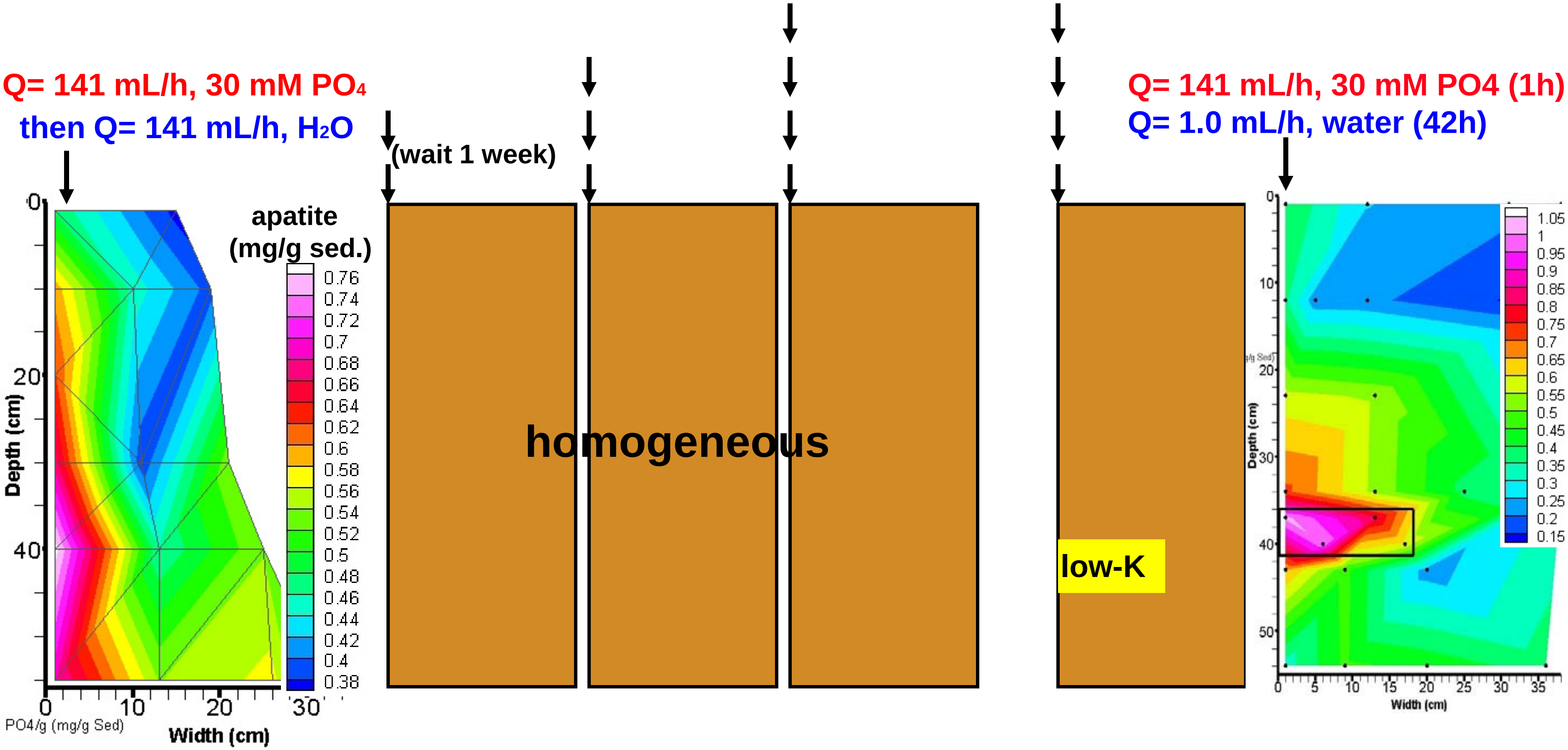




- **flow manipulation (and high residual water content) can be used to emplace apatite in low-K zones at depth**

Sequential Infiltration Events

Can repeated solution infiltrations be used to increase apatite mass?

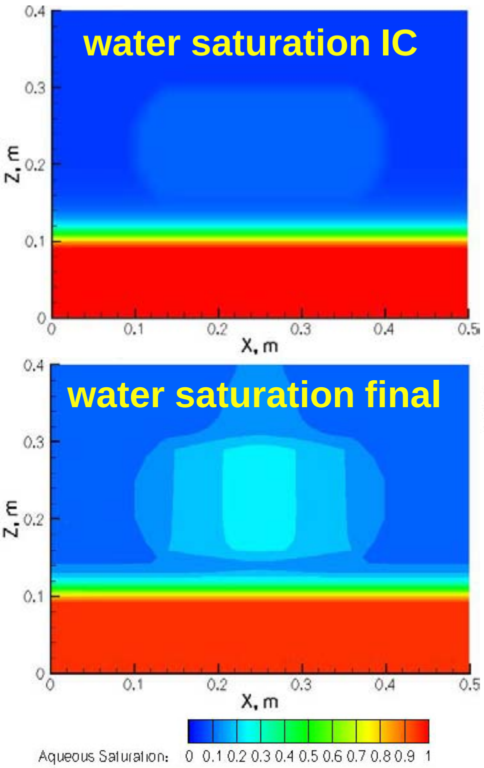


- likely less efficient for short term cycles; ppt likely nearer surface (residual water, biomass buildup)
- long term cycles better?

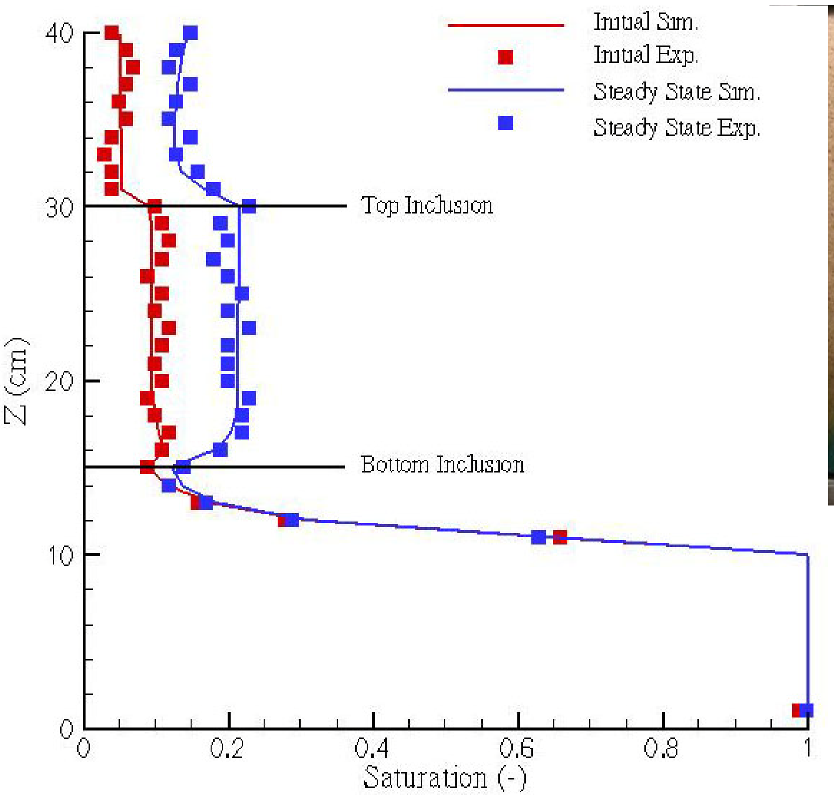
2-D Infiltration Experimental Results (EMSL)

purpose: • control water content and residence time in and under low-K lens
• measure apatite precipitate spatial distribution
(PO_4 mass balance, PO_4 extraction)

Experiment Presimulation

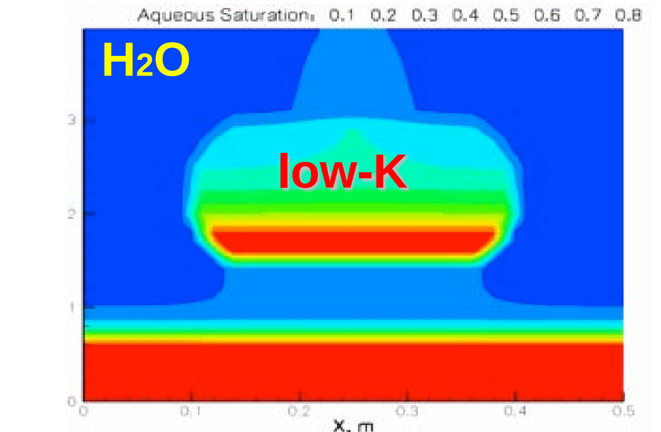


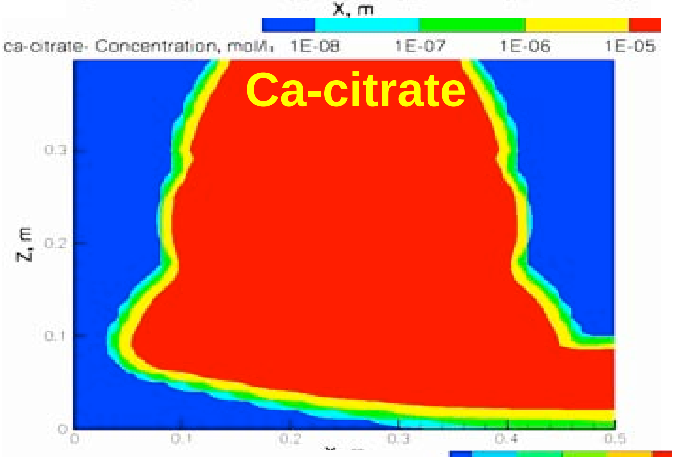
Water Content Vertically in Center (data, sim.)

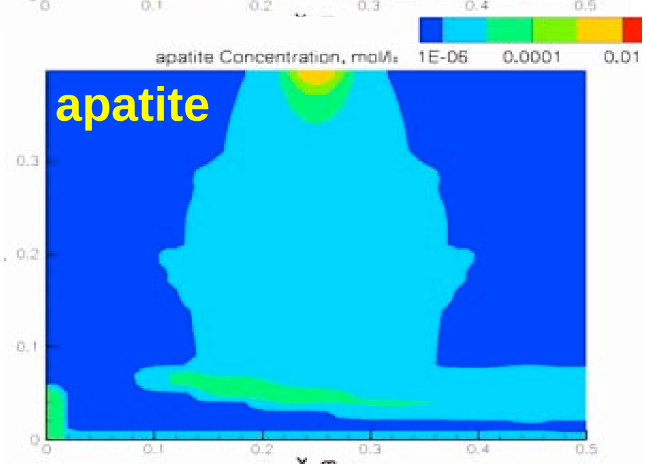


Dye Injection after Ca-citrate-PO4 Experiment

2-D Infiltration Simulations





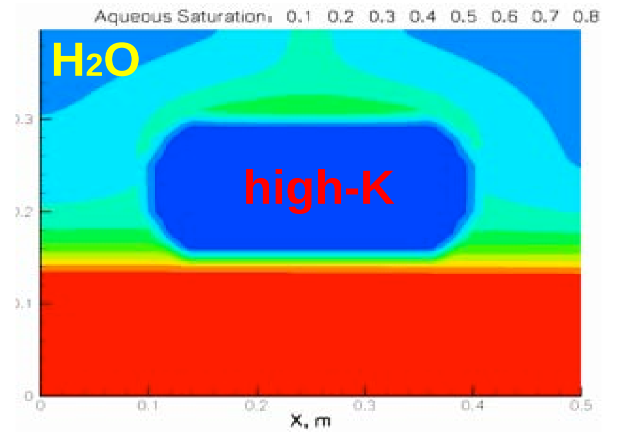


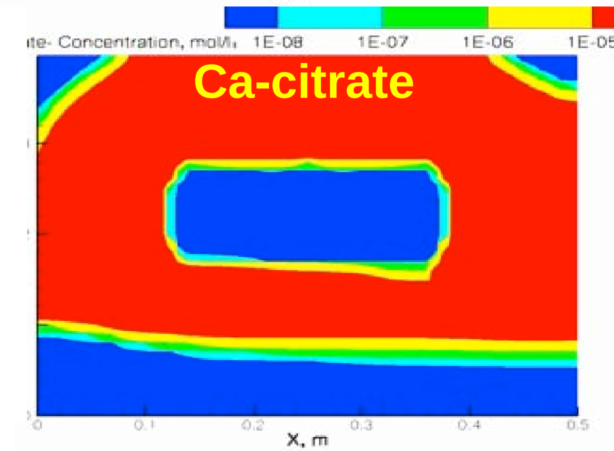
setup:

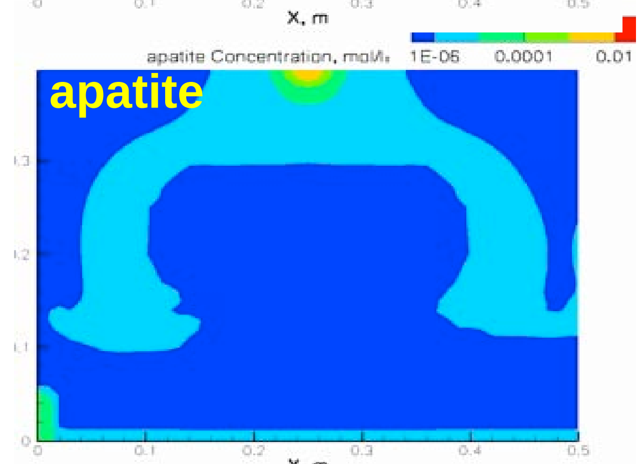
- slow infiltration rate (v = 1.6 cm/h)
- to 480 h, 120 h shown

results:

- more apatite in low-K zones (less in high-K)
- more rapid infiltration results in deeper apatite ppt, as shown in experiments



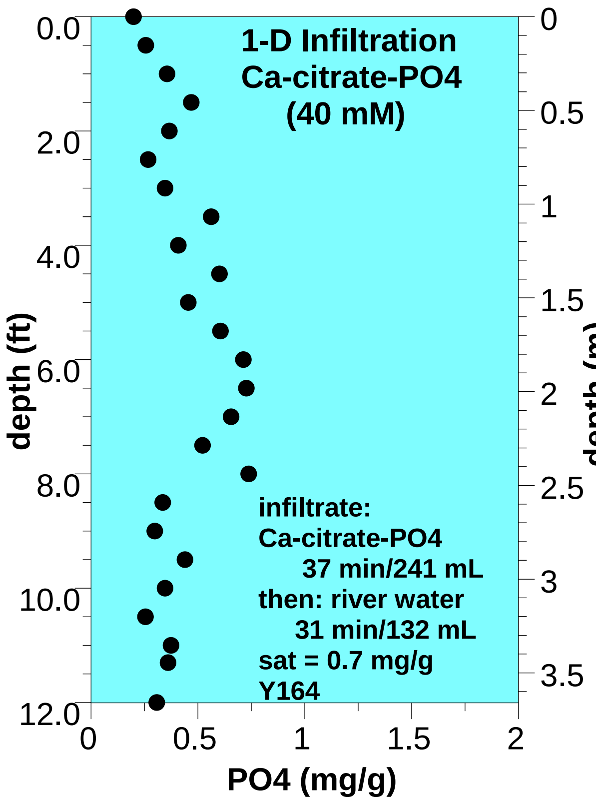


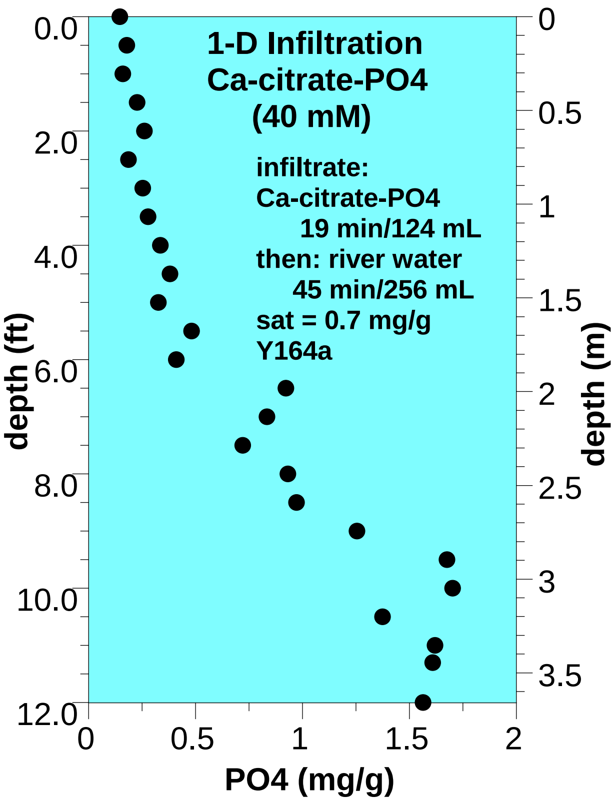


Large Lab-Scale Infiltration

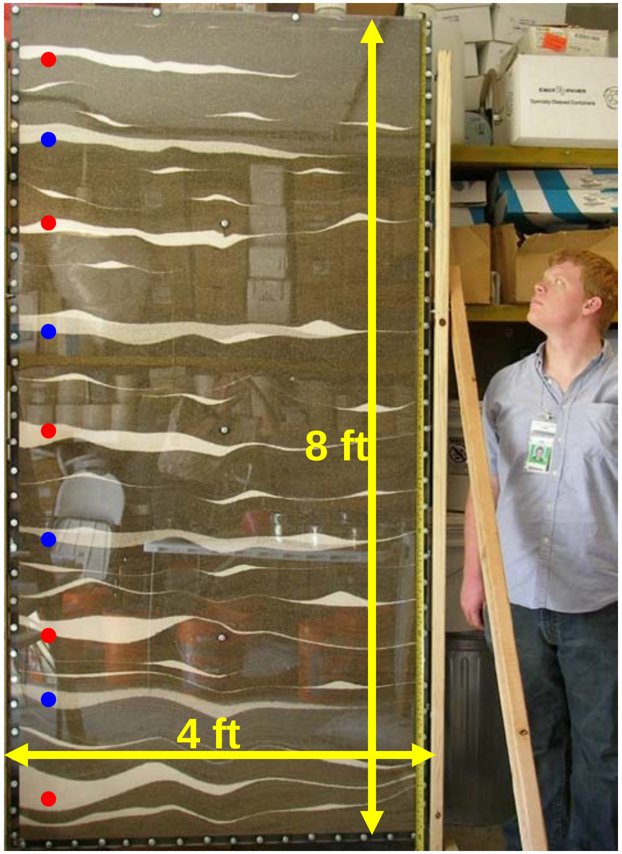
• can the infiltration strategy from < 1 m 2-D lab systems apply at 3-5m field scale, or will there be scale effects such as limited infiltration rate?

1-D infiltration (12 ft):





2-D system (8 ft):



same flow as smaller scale:
- too much PO_4 sorption limited depth

faster infiltration rate:
- most PO_4 in lower half

matrix = Hanford Fm < 4 mm
• = low-K layer, 0.2 mm
• = high-K layer, 1.2 mm

Microbial Transport and Apatite Formation

Fluor funded task

- **microbes needed for citrate biodegradation**
- **microbes in river water and aquifer (primarily on sediment, 10^4 - 10^8 cells/g)**
- **small scale data indicates redistribution of microbes upon infiltration**

Questions:

- **is the ionic strength and/or injection velocity (shear force) moving microbes?**
- **are aquifer and/or river microbial population primarily degrading citrate?**
- **is a subset of the population primarily degrading citrate?**

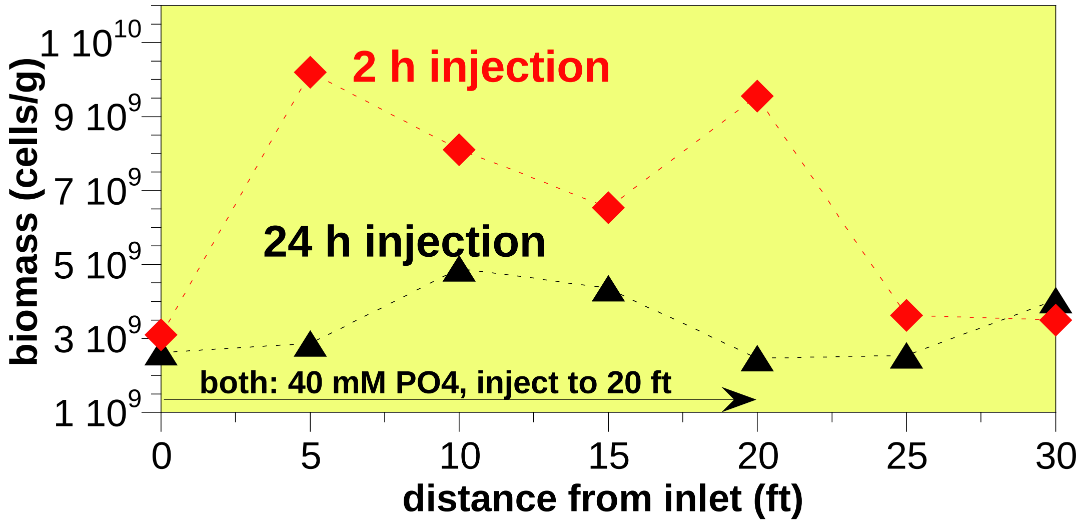
Experiments:

(5) 30-ft sediment sat. columns, (2) unsat., inject Ca-citrate-PO4 sol'n to 20 ft PO4, biomass (AODC, PLFA), PCR/DNA analysis 25 samples along length

- **ionic strength: 40 mM PO4 vs 10 mM PO4 (both 24 h injection)**
- **injection velocity 2 h versus 24 h injection (both 40 mM PO4)**
- **river versus aquifer microbe primary control:**
  - **(1) sterile sediment (only live river microbial population)**
  - **(1) sterile injection solution (live sediment microbial population)**
- **population subset: track DNA profile of population over time, distance (EBI-finded)**

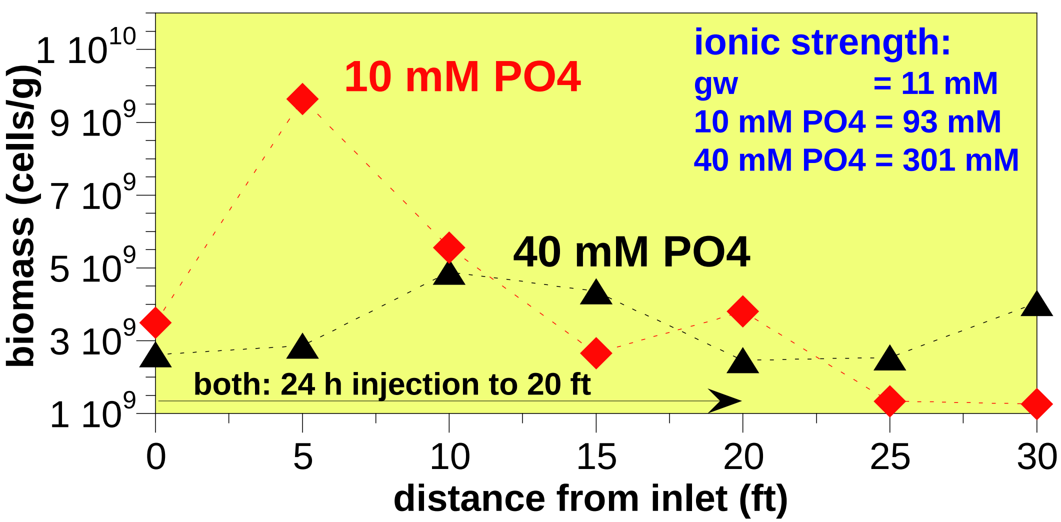
Microbial Transport Processes

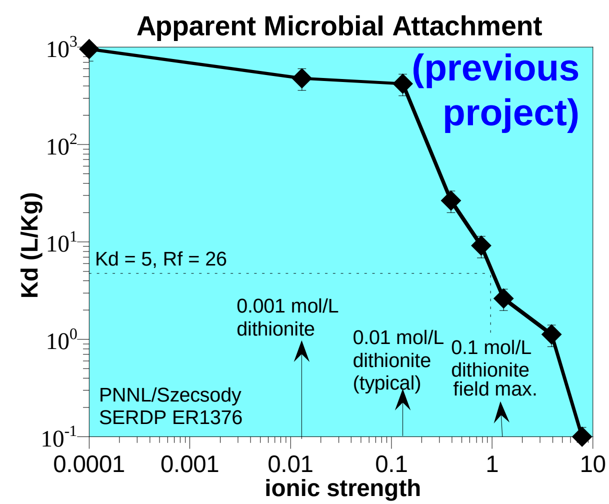
Influence of Flow Rate: high shear moving in situ microbes



biomass by AODC

Influence of Sol'n Conc.: higher ionic strength moving microbes further

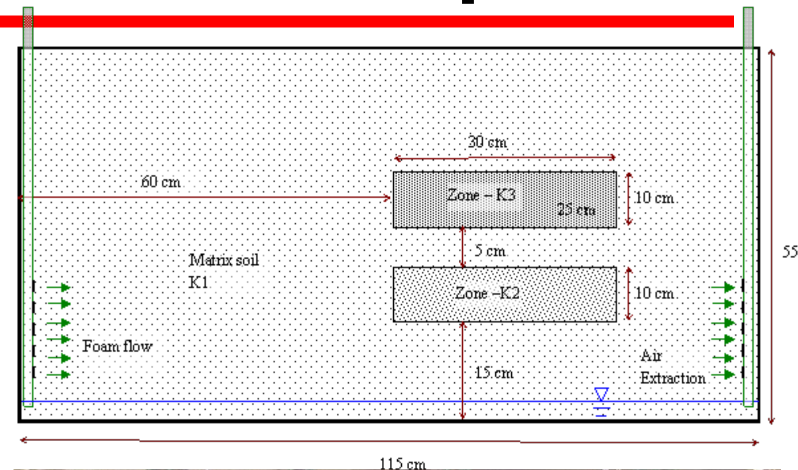


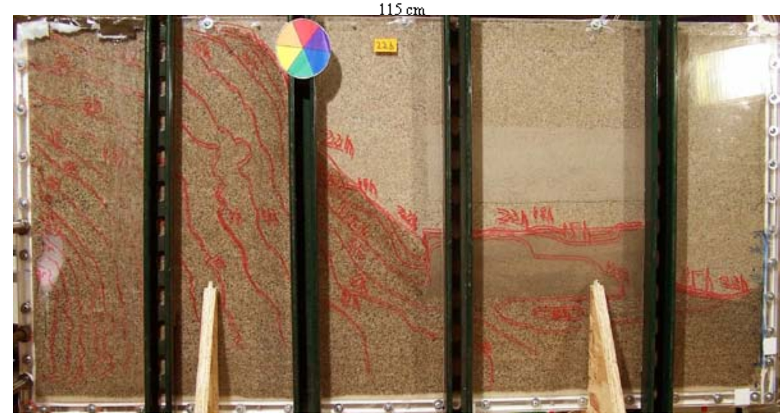


Use of Foam for Low Water Transport

- **new low-water injection technology developed by Lirong Zhong (PNNL/LDRD funding) injecting polysulfide [Battelle-business sensitive*]**

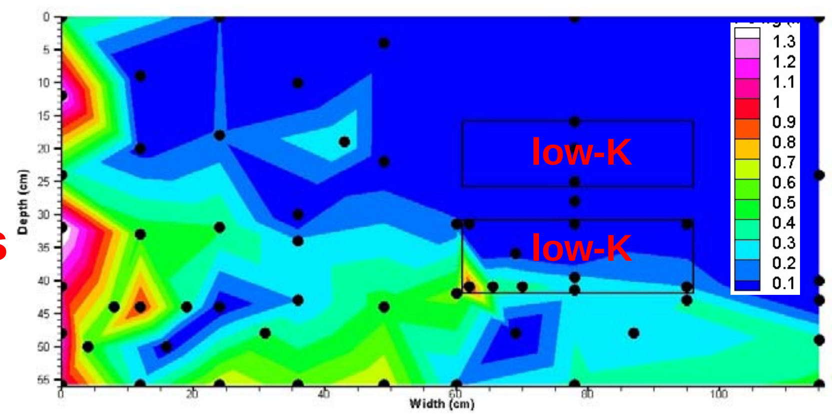
- **tested with Ca-citrate-PO_4 solution on this EM-22 project**





- **(2) 2-D injections, 1.3 m x 0.4 m**
  (2) 1-D injections, 7 m

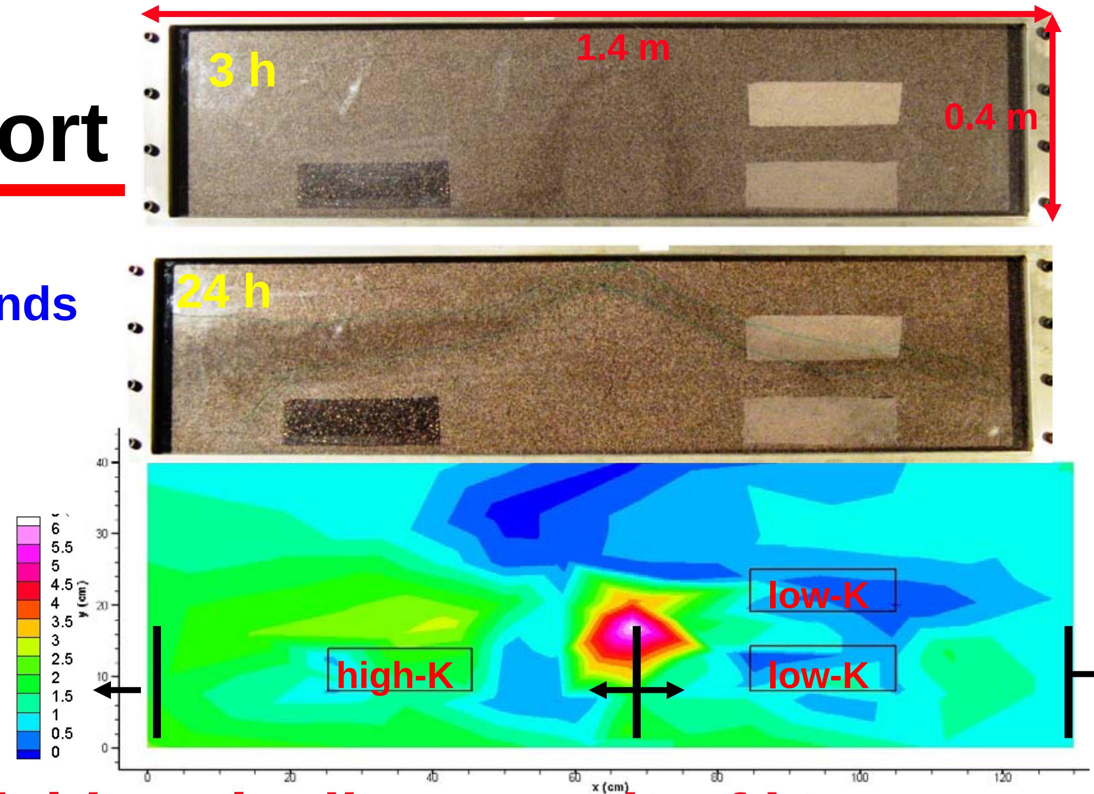
** This document contains Battelle-Proprietary Information and, since it is transmitted in advance of patent clearance, is made available in confidence under the protections of 18 USC §1905 solely for use in performance of work under contracts with the U.S. Department of Energy. This document is not to be published nor its contents otherwise disseminated or used for purposes other than specified above before patent approval for such release or use has been secured, upon request, from Intellectual Property Legal Services, Pacific Northwest National Laboratory, Richland, Washington 99352.*



- **might be useful to inject into 100N wells at low river stage to treat high-K zones of the vadose zone**

Use of Foam for Low Water Transport

- 10 pore volumes foam injected
- center injection, withdraw both ends



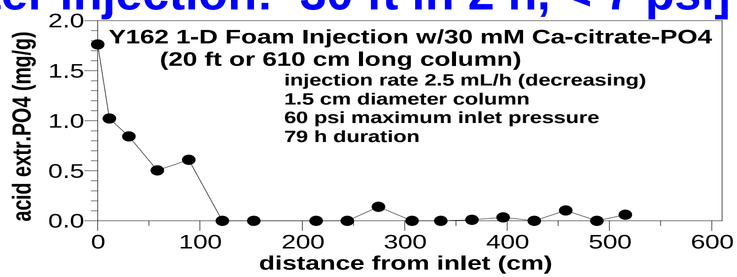
- more flow to high-K zones
- high apatite ppt near high-K (no apatite in low-K)
- 1% Na (very high ionic strength)

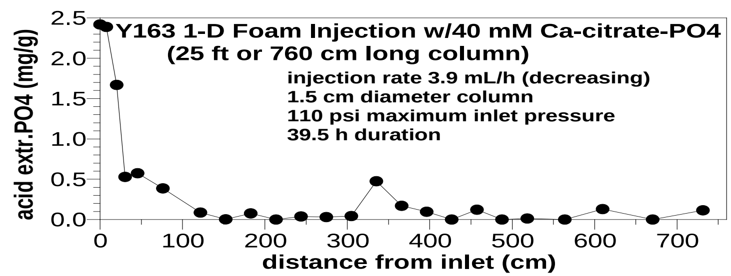
Can foam be injected to field-scale distances (25 ft)?

- sand/silt (< 4 mm); moisture to 20 ft, PO_4 to 4 ft (60 psi, 79 h)

[water injection: 30 ft in 2 h, < 7 psi]

- sand (0.5-4 mm); moisture to 20 ft, PO_4 to 3 ft (110 psi, 40 h)

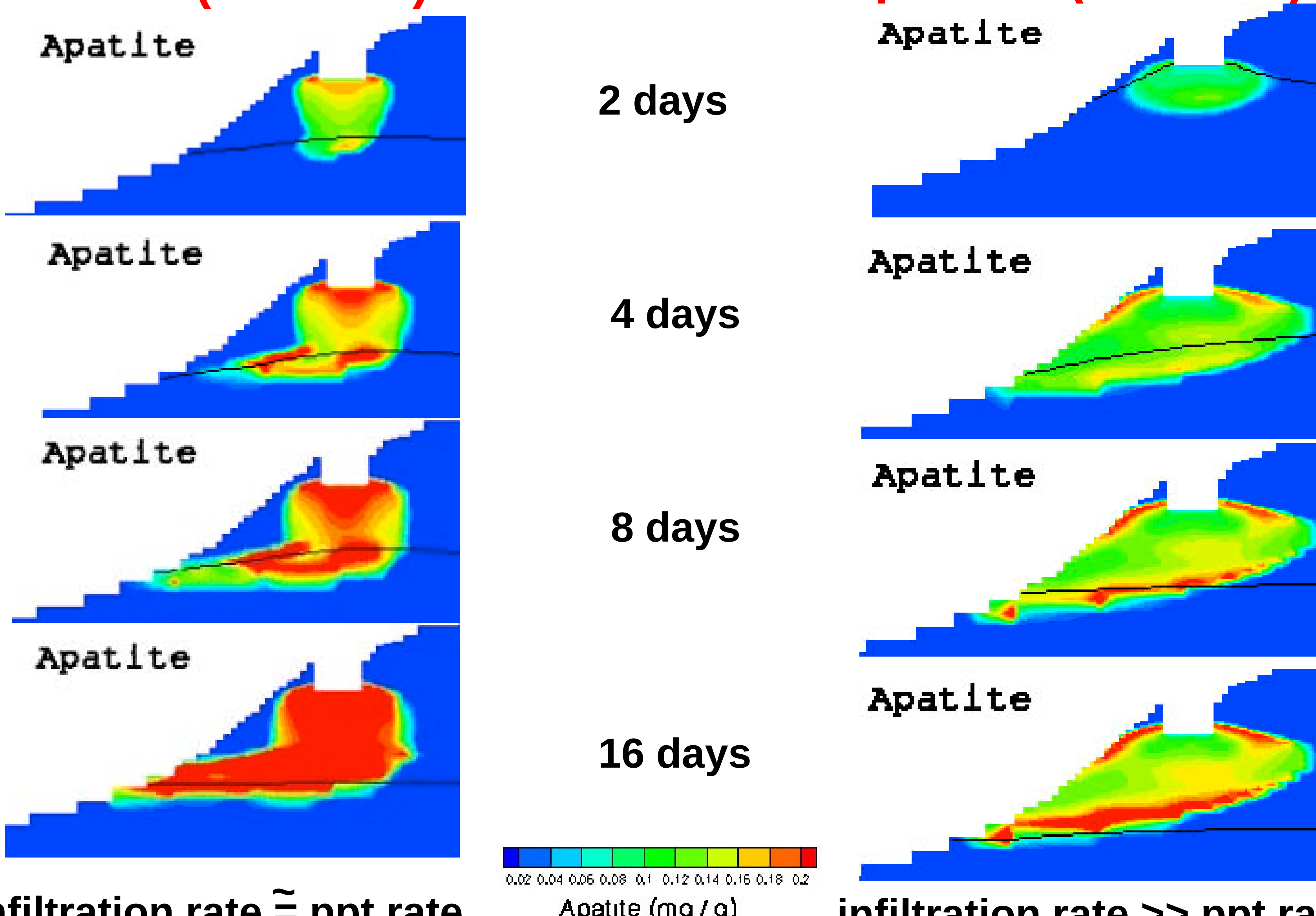




conclusions: foam advection too slow (and too much Na) for aq. reactant; may be appropriate for solid phase injection

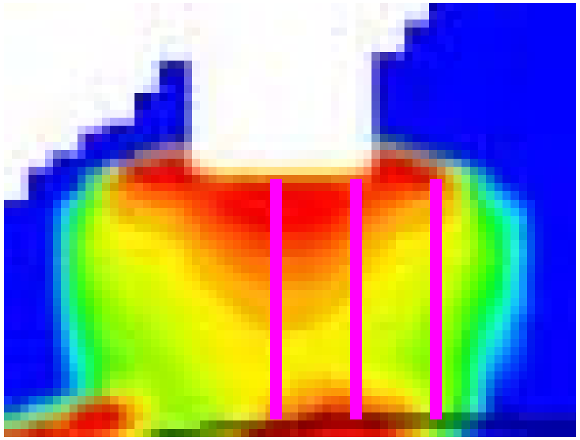
Field-Scale Simulation of Apatite Processes

trickle (0.1 cm/h): | **ponded (1.0 cm/h):**



infiltration rate ≅ ppt rate | **infiltration rate >> ppt rate**

Preliminary Field Infiltration Tests



FY08:

- **water infiltration tests in road fill (trenches)**
  [can we infiltrate in the road fill?]

FY09:

- **planned line infiltration in trench**
- **6 neutron access holes (in line, perpendicular)**
- **6 sets of nested:**
  lysimeters for sampling,
  with EC electrodes for cross borehole resistivity

Summary

processes

- Apatite forms under unsaturated conditions; controlled by:
  (a) PO_4 adsorption (hours), (b) citrate biodeg. (10s hours), (c) infiltration rate
- citrate biodeg. relatively constant even with biomass variation
  microbial transport study in progress
- Sr-90 is incorporated into apatite slowly: 5.5 to 16 month half-life
- 3.4 mg apatite/g sed. sufficient for sequestering Sr-90 for 300 yrs

w/unsat. transport

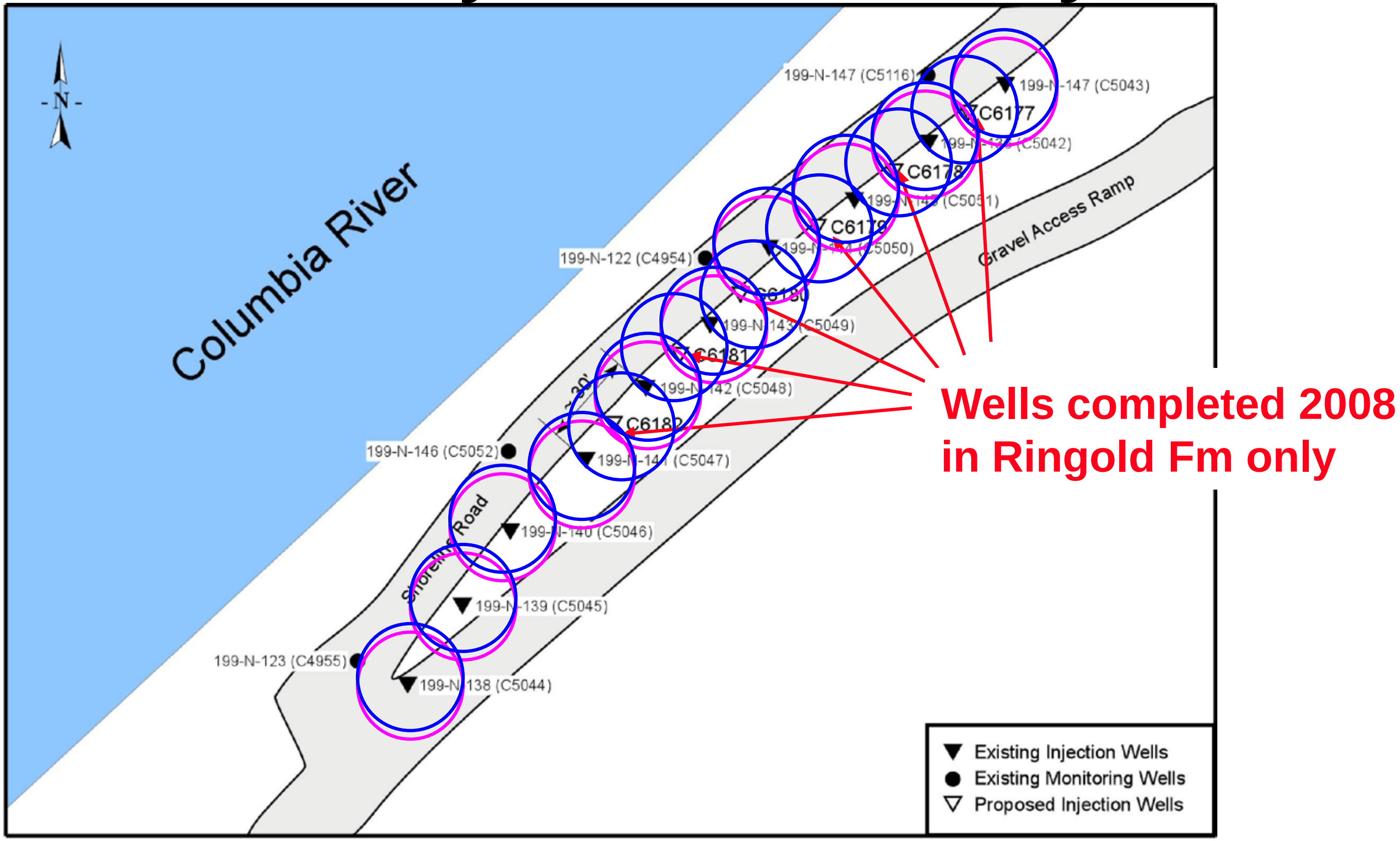
- If infiltration residence time >> biodeg. half-life (50-150 h)
  resulted in deeper/more uniform apatite area coverage
- Rapid PO_4 solution infiltration followed by slow water infiltration
  can emplace apatite ppt at depth and within low-K zones
- Upscaling shows need for modification of infiltration strategy
- Further develop infiltration strategy based on experimental and
  simulation results for a wider range of field conditions

foam

- Foam use for Ca-citrate-PO4 not efficient and targets high-K zones;
  may be useful for advecting in particulate reactants

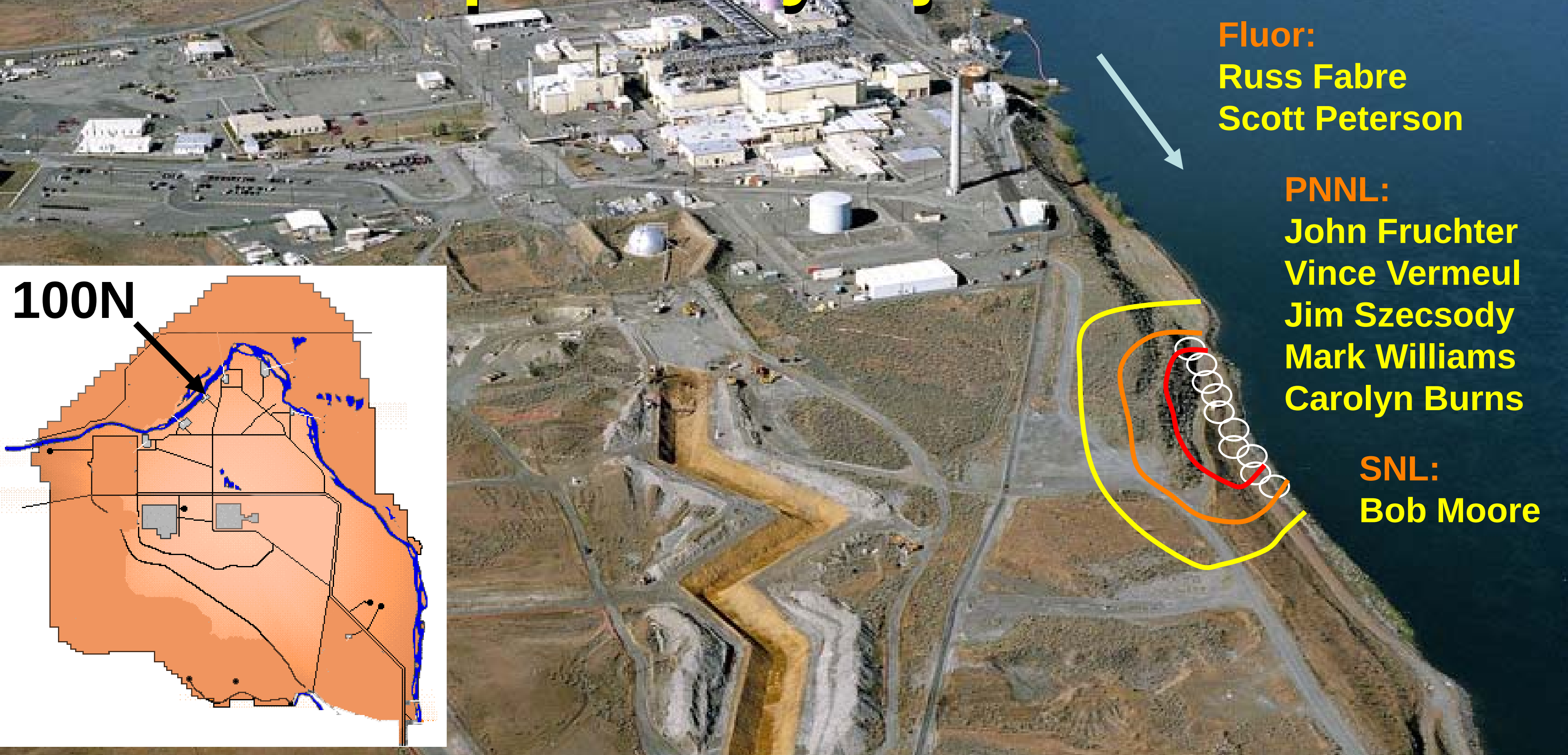
backup slides

100N Injection Summary



- 2005-07 (18) low conc. injections (10 mM PO4)
- 2008 (16) high conc. injections (40 mM PO4)

Sr-90 Immobilization by Apatite: Emplaced by Injection



- Sr-90 in 100N sediments leach into the Columbia River
- Sr adsorbs strongly, so only Sr-90 in near-river sediments would reach river before natural radioactive decay

Sequential Ca-Citrate-PO_4 Injections for Sr-90 Remediation

purpose: inject needed PO_4, but minimize short-term Sr-90 peak

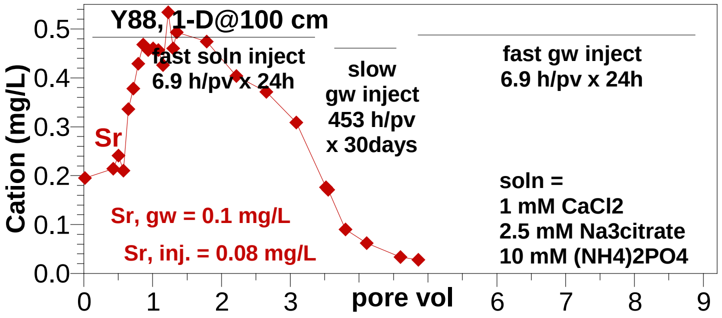
Inject Low Conc. ⇨ **Wait 1 year** ⇨ **Inject High Conc.**

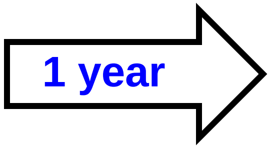
Inject Low Conc.
- 1 mM Ca
- 2.5 mM Citrate
- 10 mM PO_4

Wait 1 year
for ~50% of Sr-90 in inj. zone to be incorporated into apatite

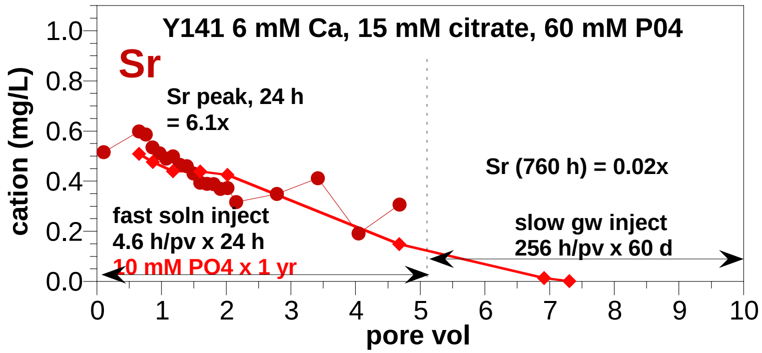
Inject High Conc.
- 6 mM Ca
- 15 mM Citrate
- 60 mM PO_4

Lab Exp. 1/2007





Lab Exp. 1/2008 (07 sed.)



Lab Results:
- Sr peak (24 h) = 5x
- Sr (30 days) = 0.28x

(field peak, 24 h = 4x)

Lab Results:
- Sr peak (24 h) = 6-11x
- Sr (30 days) = 0.02x

Lab Tests for High Concentration Ca-Citrate-PO_4 Injections

Baseline (no low conc. pretreatment)

- 3 mM Ca, 7.5 mM, 30 mM PO4

1. Inject Ca-Citrate-PO_4 (1 year after: 1 mM Ca, 2.5 mM cit., 10 mM PO4 treatment)

- same stoichiometry as field injections #3 to #18 (but higher conc.)
- Ca = 2, 3, 4, 6 mM
- Citrate = 5, 7.5, 10, 15 mM
- PO_4 = 20, 30, 40, 60 mM

2. Inject Citrate-PO_4 (no Ca)(1 yr after: 1mM Ca, 2.5mM cit., 10mM PO4 treatment))

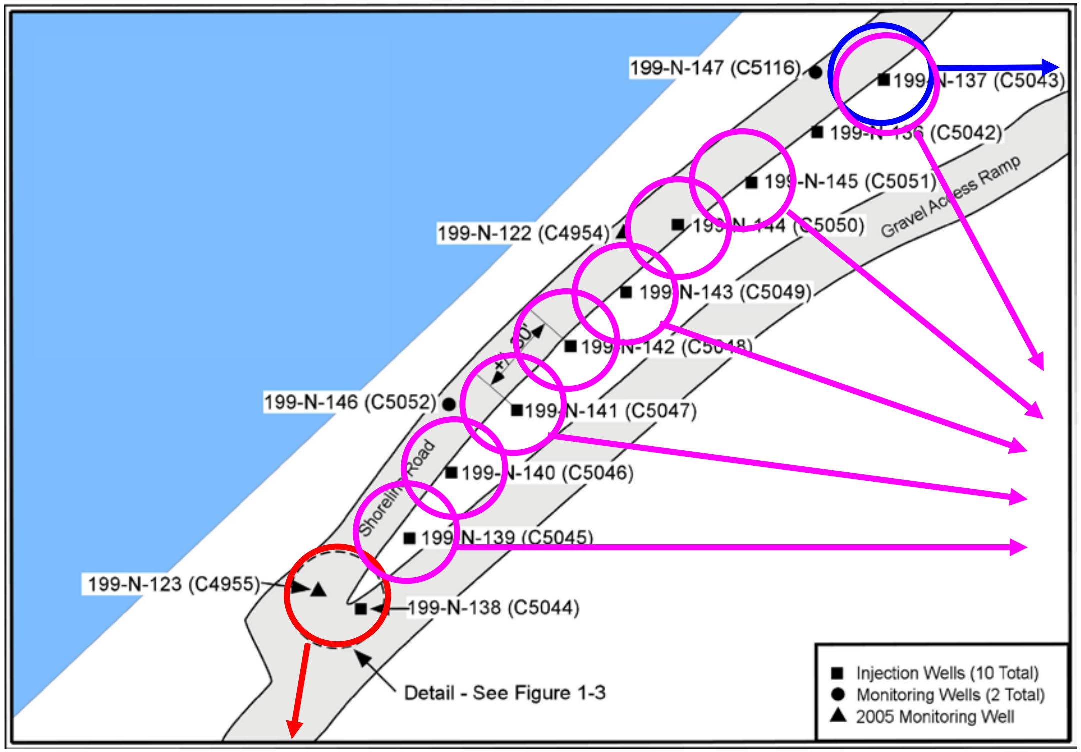
- Ca-poor solution will utilize more Sr and Ca in sediment
- PO_4 = 30 mM (all)
- Na-Citrate = 2.5, 7.5, 22 mM

3. Inject Ca-Citrate-PO_4 + F- (1 yr after: 1mM Ca, 2.5mM cit., 10mM PO4 treatment))

- fluorine (substitutes for OH- in apatite) increases precipitation rate
- Ca = 3 mM, Citrate = 7.5 mM, PO_4 = 30 mM (all)
- F- = 0, 0.21 (drinking water conc.), 2.1 mM

Each Experiment: 1-D column, 24 h solution injection, then 60 day groundwater inj.
measure: Ca, Na, K, Sr, PO4, citrate, EC
objective: peak Sr (during sol'n inj.), Sr (60 days)

100N: Low Conc. Injections



Pilot Test #2:

- 60,000 gal injection 9/27/06
- 2 mM Ca, 5 mM citrate, 2.4 mM PO4
- low river stage injection
- Treat Ringold Fm
  - good IW efficiency; Hanf./Ring. K contrast 7x
- Average increase in Sr-90 3.3x (range 0 to 6.2x)

Implementation #3 - #18:

- 140,000 gal injection/60 h, 2/07 to 7/07
- 1 mM Ca, 2.5 mM citrate, 10 mM PO4
- high and low river stage injection
- Treat Hanford and Ringold Fm
- average Sr-90 increase 2.5x (days); then lower than baseline (months)

Pilot Test #1:

- 96,000 gal injection 5/31/06
- 4 mM Ca, 10 mM citrate, 2.4 mM PO4
- High river stage injection
- Treat Hanford and Ringold Fm
  - Well inefficiency; Hanford/Ringold K contrast 2x
- Average increase in Sr-90 10.5x (range 0 to 25x)

- injection strategy (chemical composition, flow rate) modified based on field data and simulations to more efficiently precipitate apatite

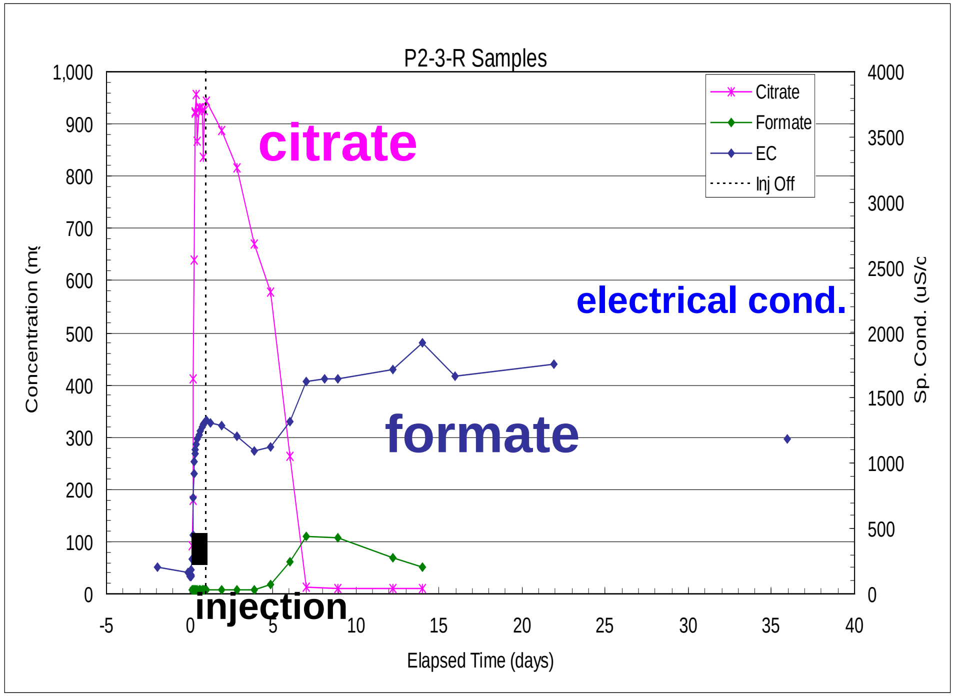
Injection Monitoring Data

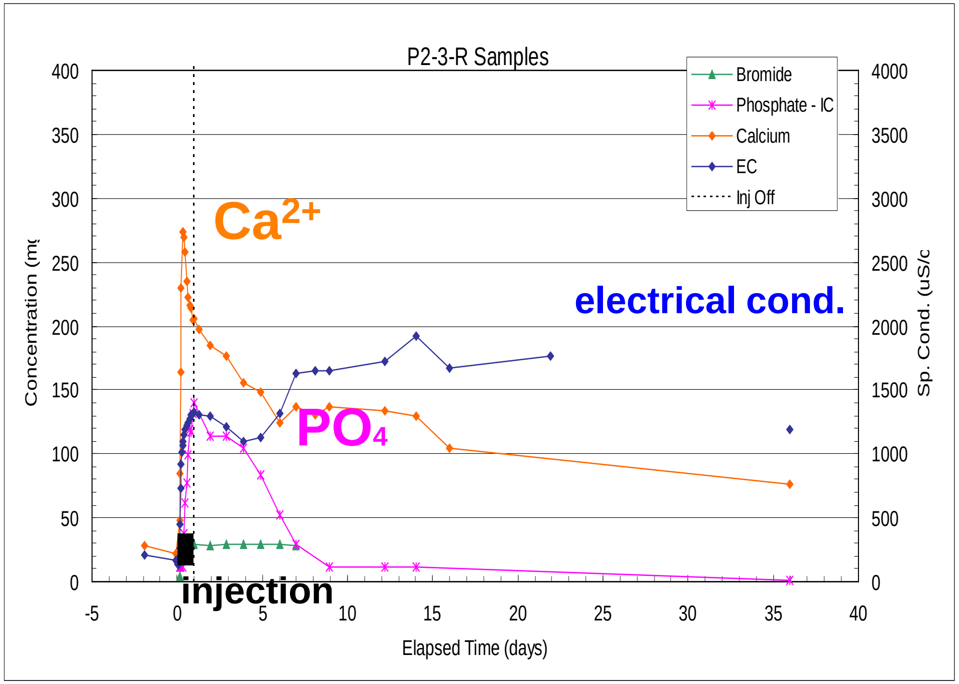
Summary based on results from Pilot test #1 and #2

Apatite Solution Precipitation

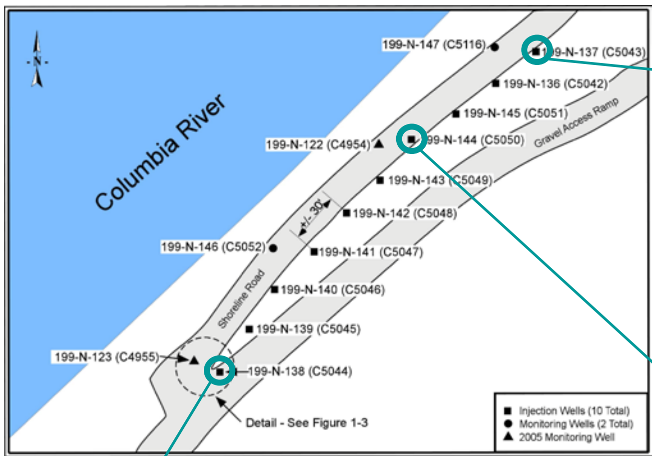
- apatite-forming precursors distributed in the subsurface
- Ca and PO_4 decrease during the reaction period; indicative of precipitation
- rapid microbial activity indicated (citrate ↓, formate ↑, O_2↓, ORP ↓)

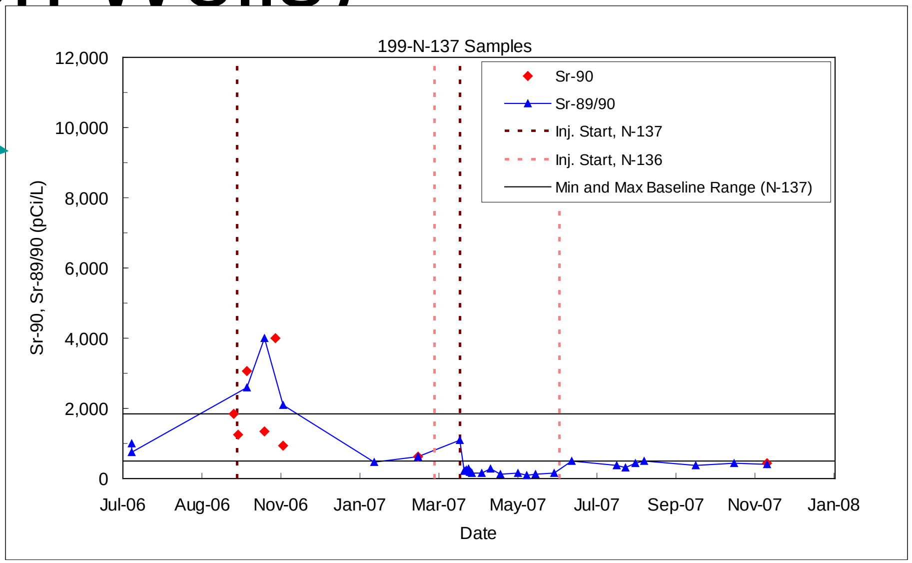
Injection data: Test #2, well P2-3R, radial dist. = 10 ft

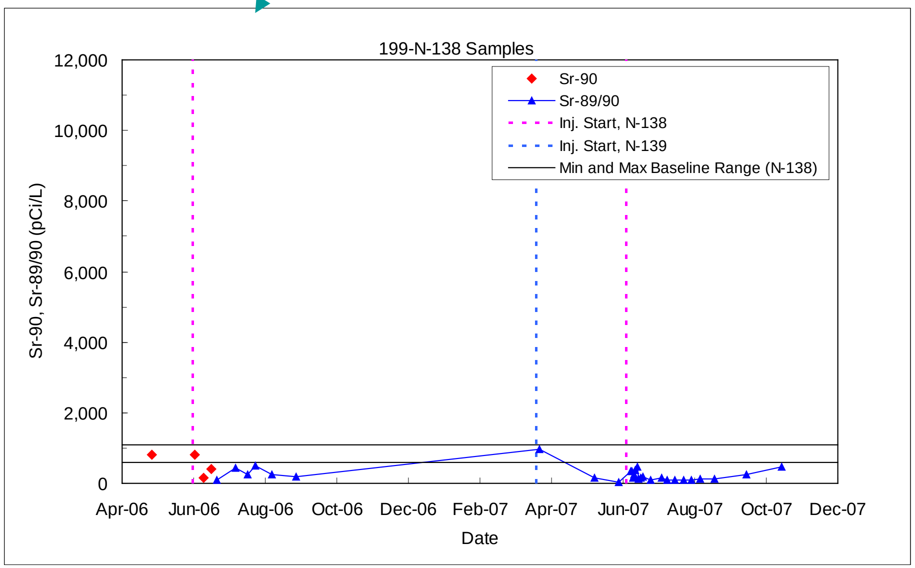


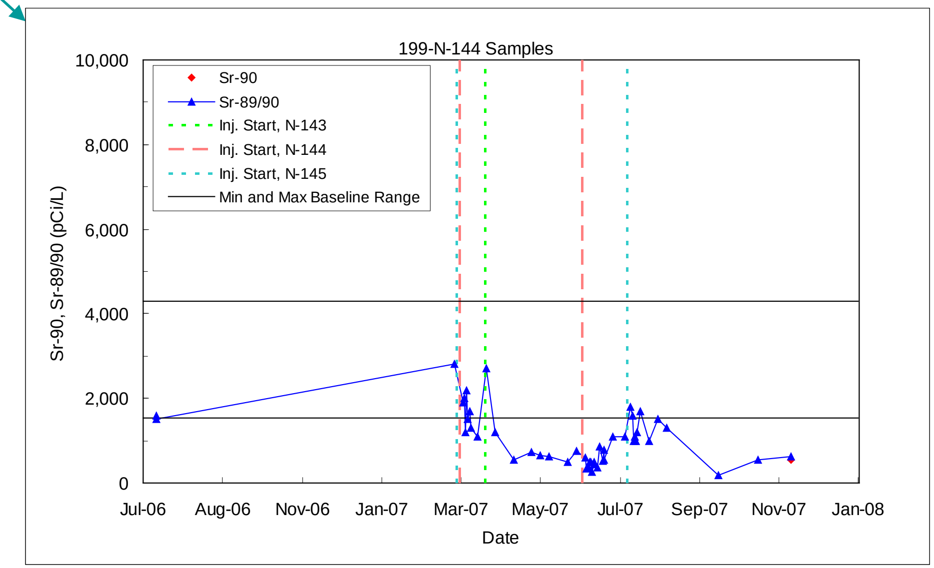


^{90}Sr Performance Monitoring (Injection Wells)

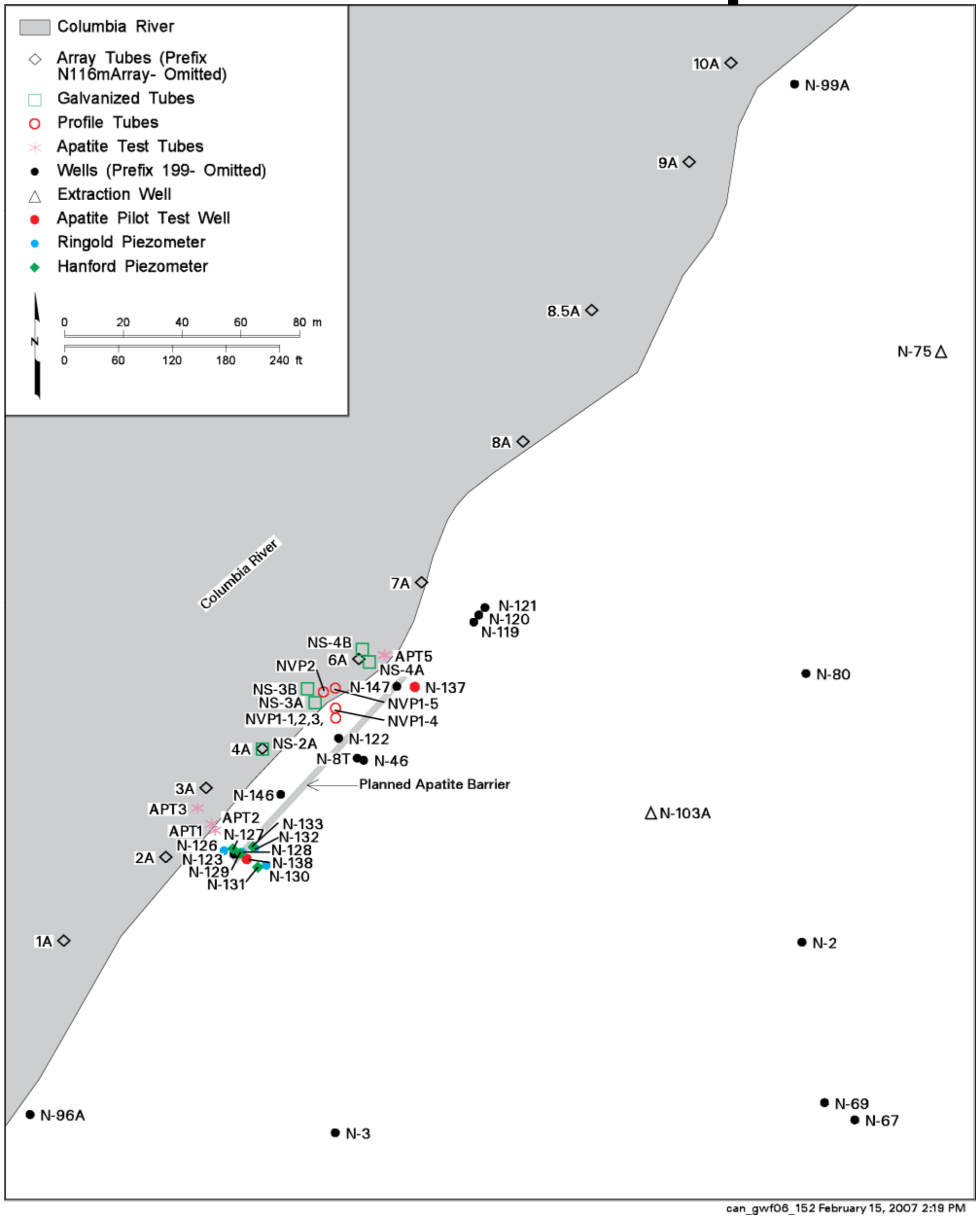


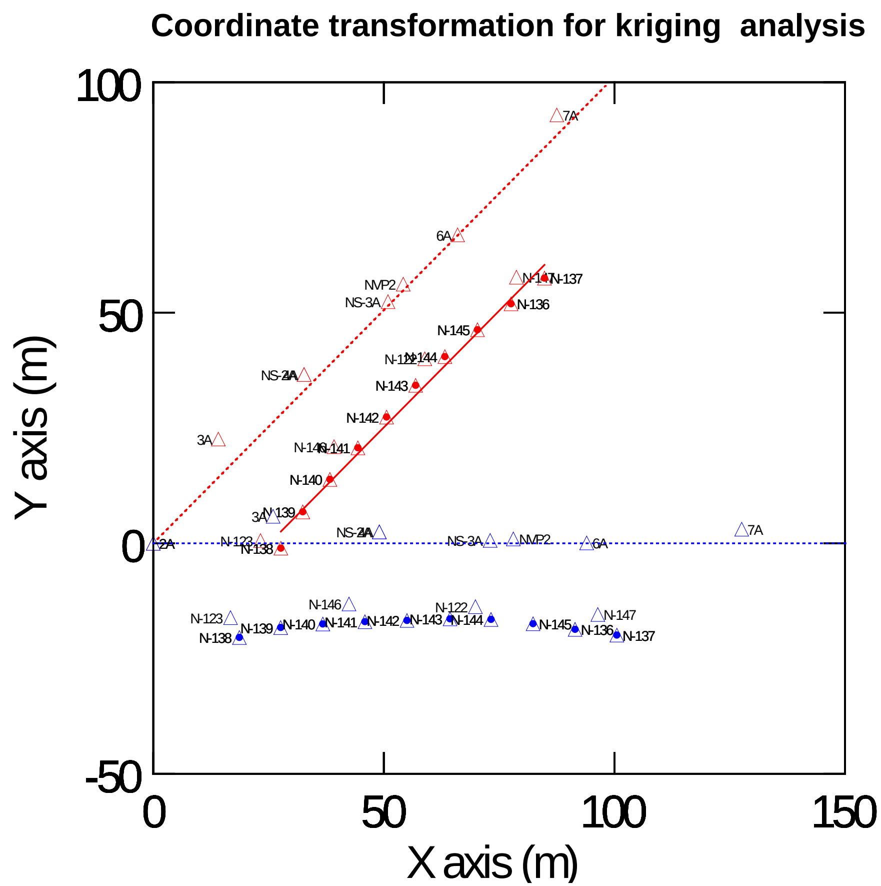




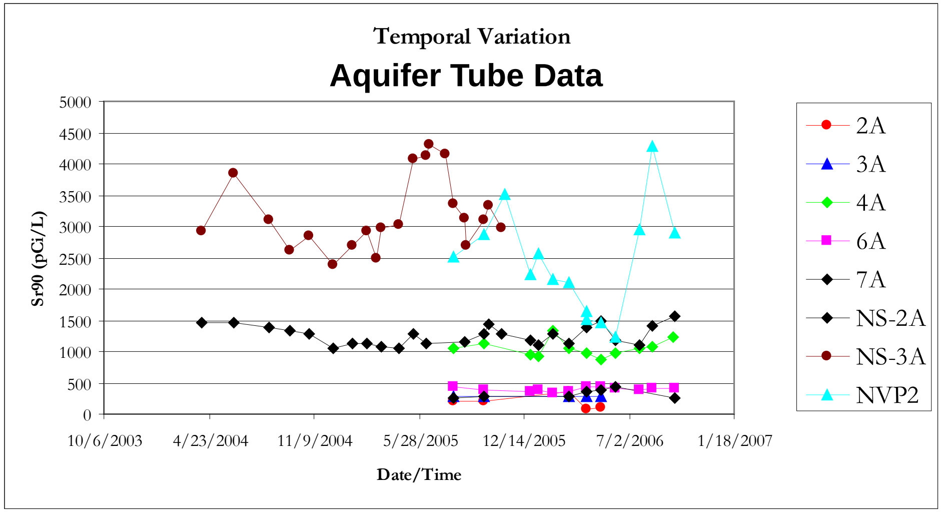


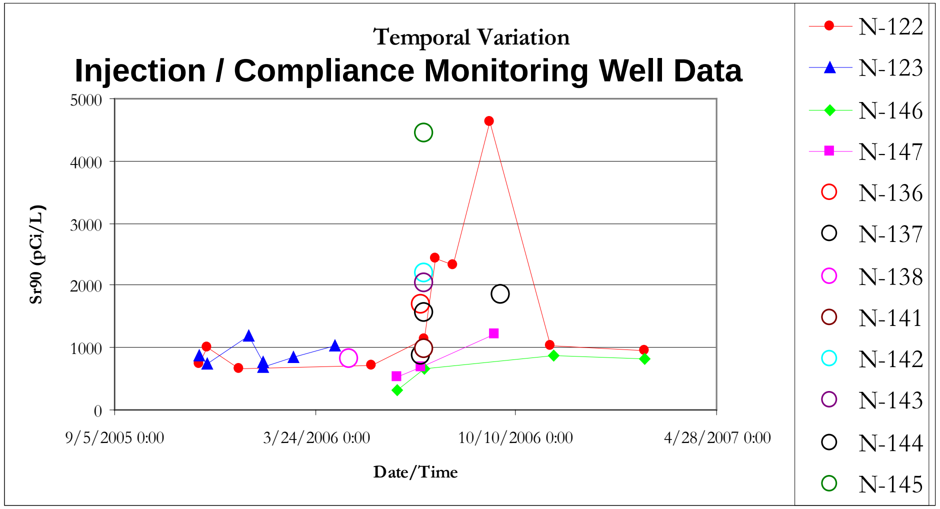
Well and Aquifer Tube Locations

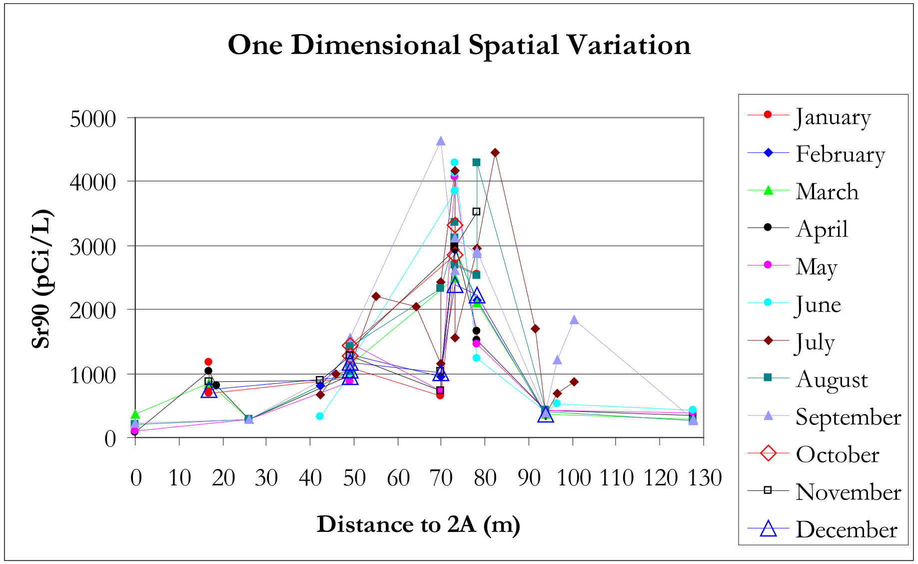




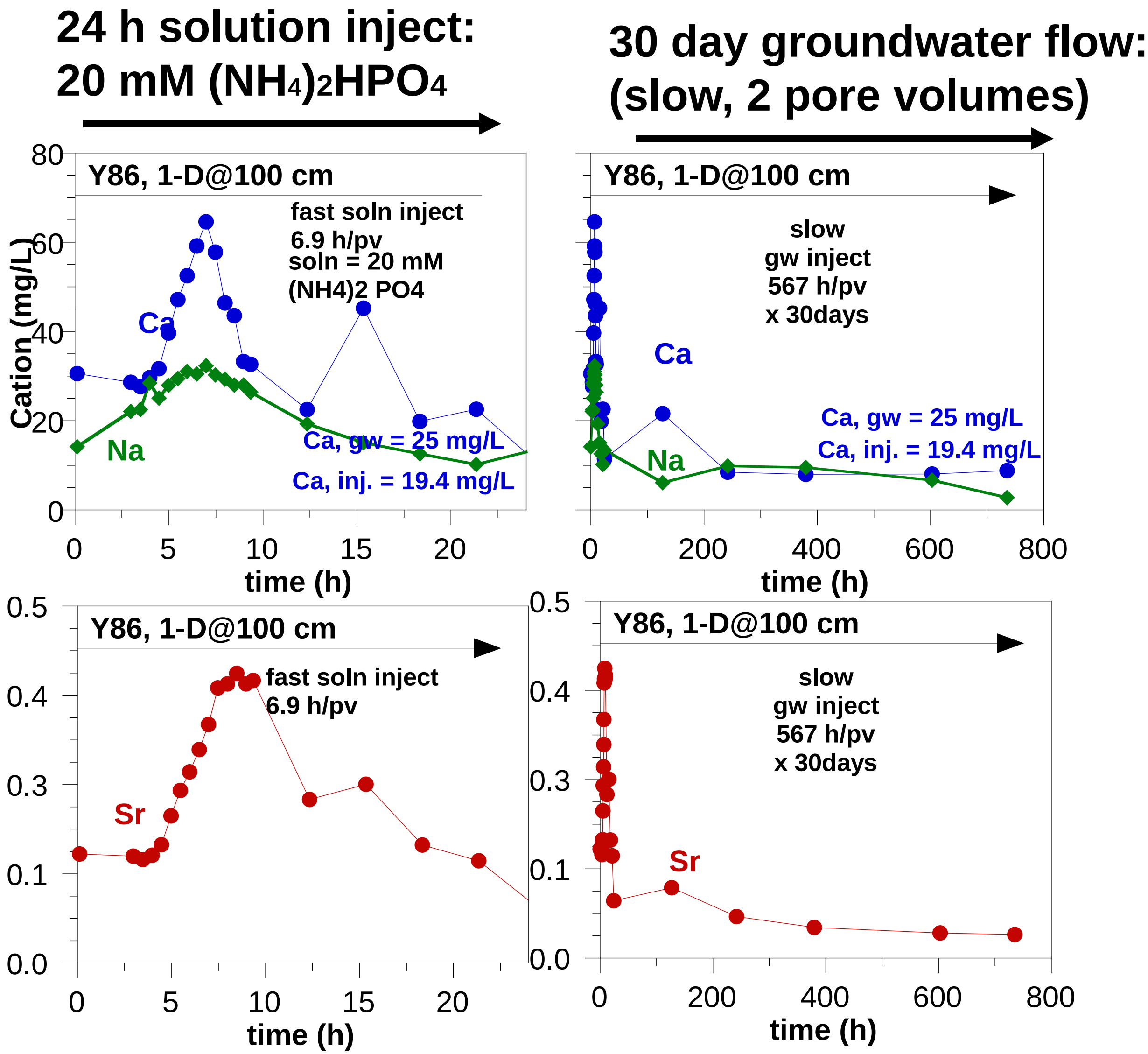
Temporal and Spatial variability in Baseline ^{90}Sr Concentrations







Sr During Solution Injection/Infiltration

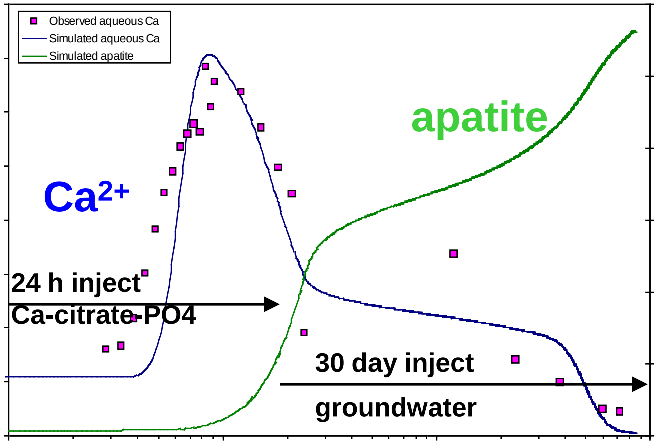


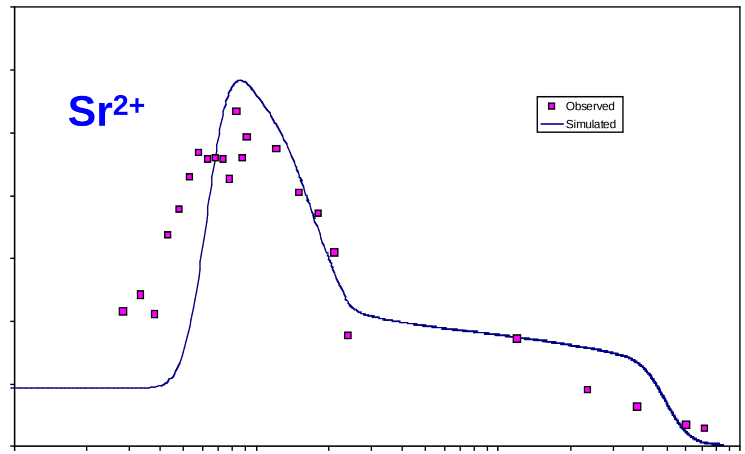
• short term Sr-90 aq increase • long-term Sr-90 aq decrease

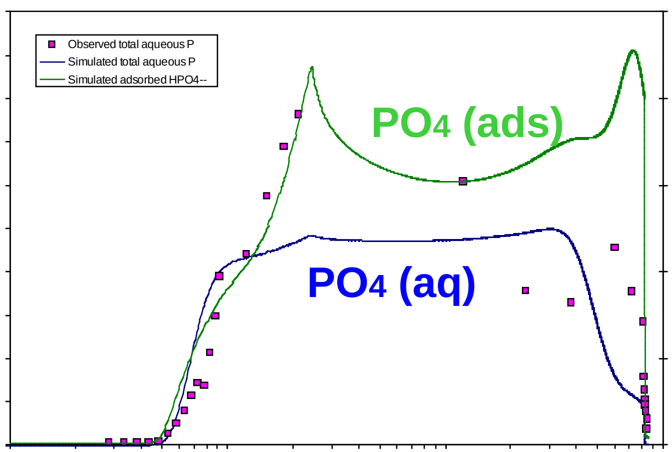
Reactive Transport Simulation (Task 4)

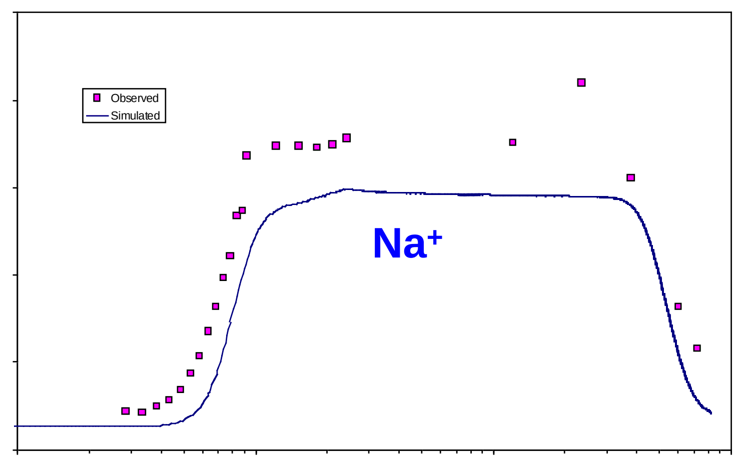
system: 1-D sat., 24 h Inject Ca-citrate-PO_4, 30 day groundwater inject

model: 3-D, saturated/unsaturated, 42 reactions, 51 species







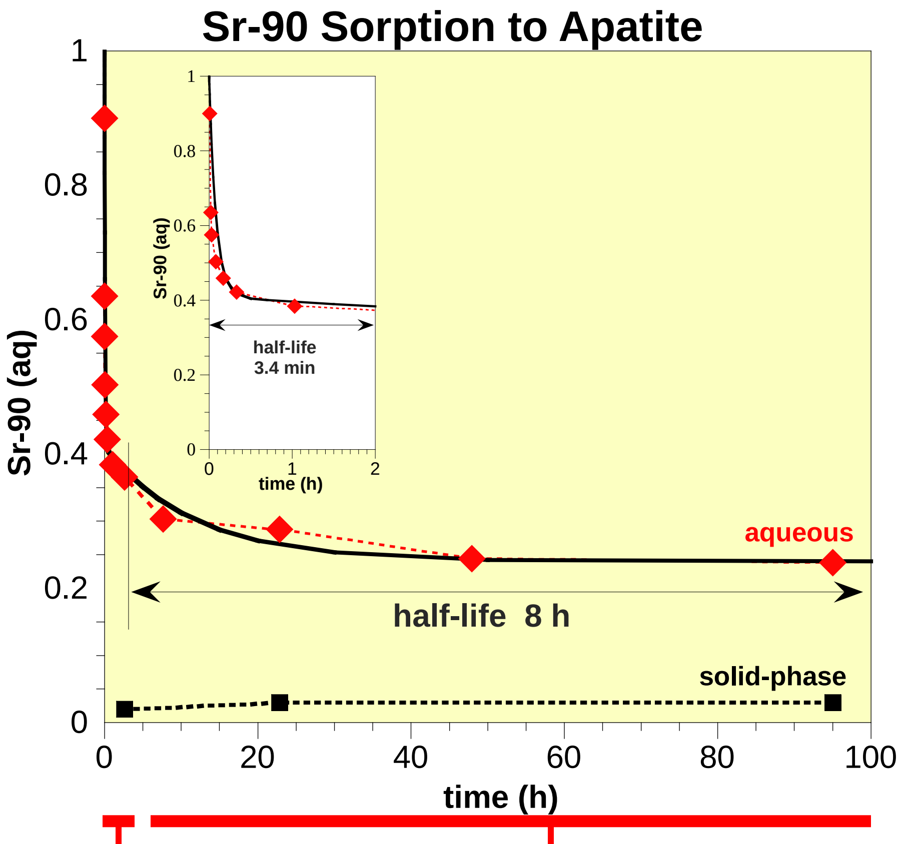


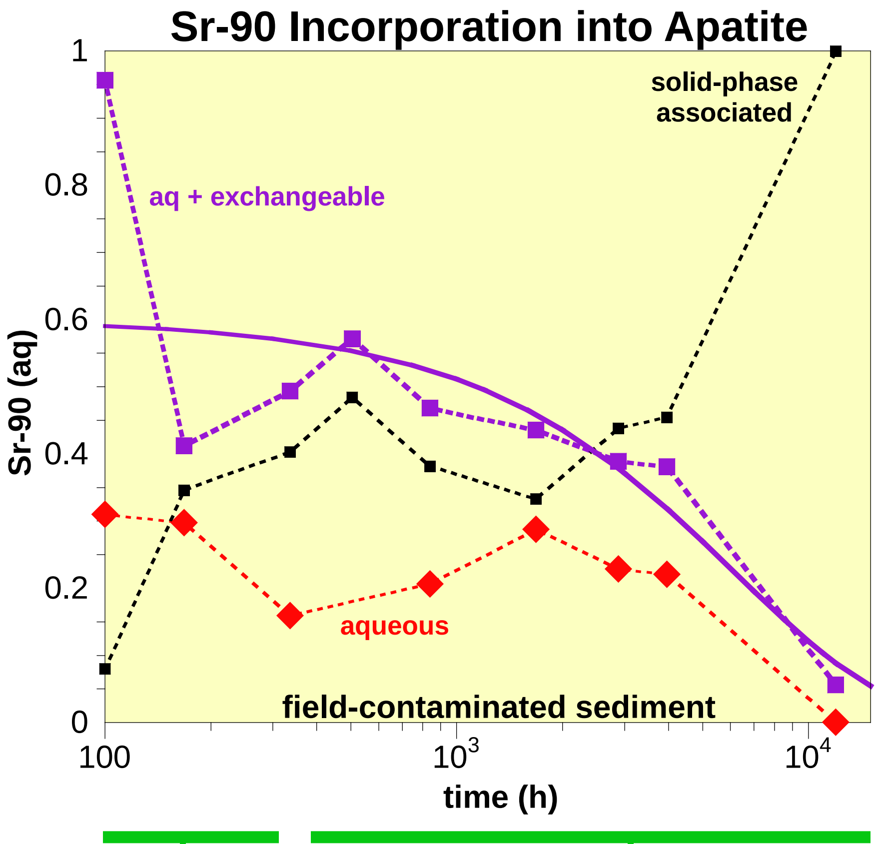
simulating major changes (many ion exch. controlled):

- injection desorbing Ca^{2+}, Sr^{2+}(8 h)
- apatite forming (>10 h) removing Ca^{2+}
- more apatite forms when Na^+ desorbs (800 h)

Task 3: Sr-90 Incorporation into Apatite

objective: determine mechanism and rate of Sr-90 adsorption and permanent incorporation into apatite





0 - 2 h: sorb by ion exch.

2 - 100 h: sorb + diffusion into porous particles

100 - 300 h: apatite ppt w/some Sr-90

72-180 d half-life: Sr-90 incorporated into apatite

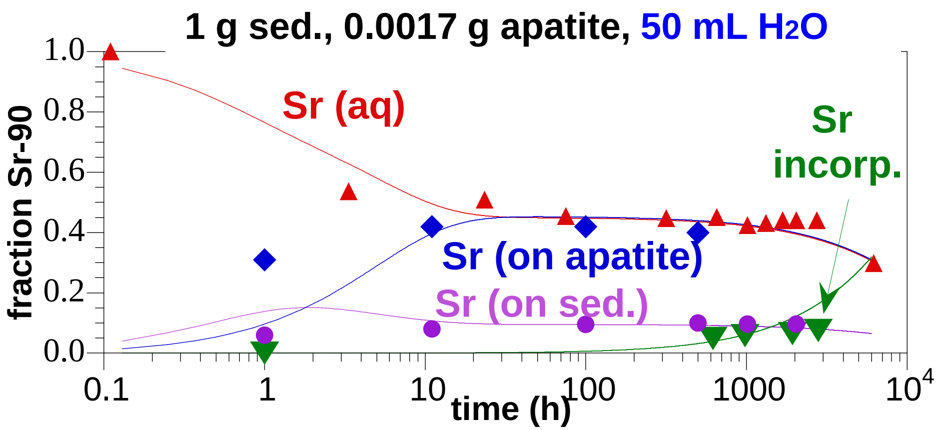
Sr-90/apatite: $K_d = 1370 \pm 429\ cm^3/g$

Sr-90/sediment: $K_d = 24.8 \pm 0.4\ cm^3/g$

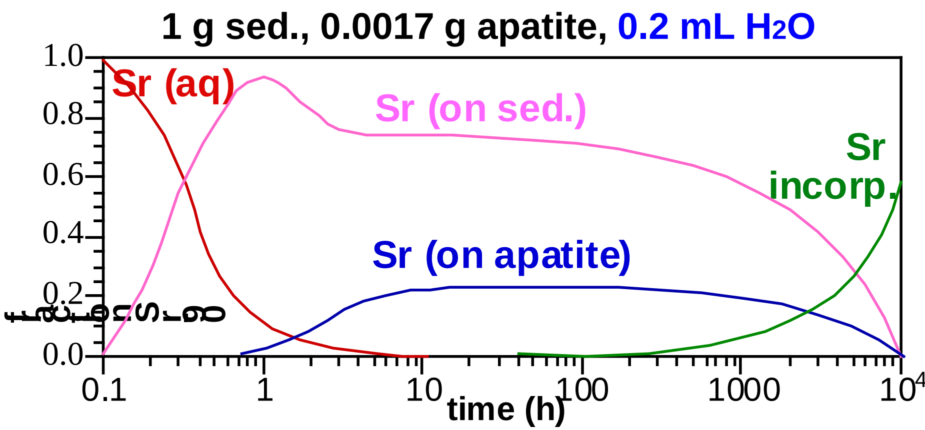
Sr-90 Incorporation: Scaleup to Field

model: Ca-Na-Sr ion exchange to sediment and apatite, Sr incorporation

Lab System



Field System (20% porosity)



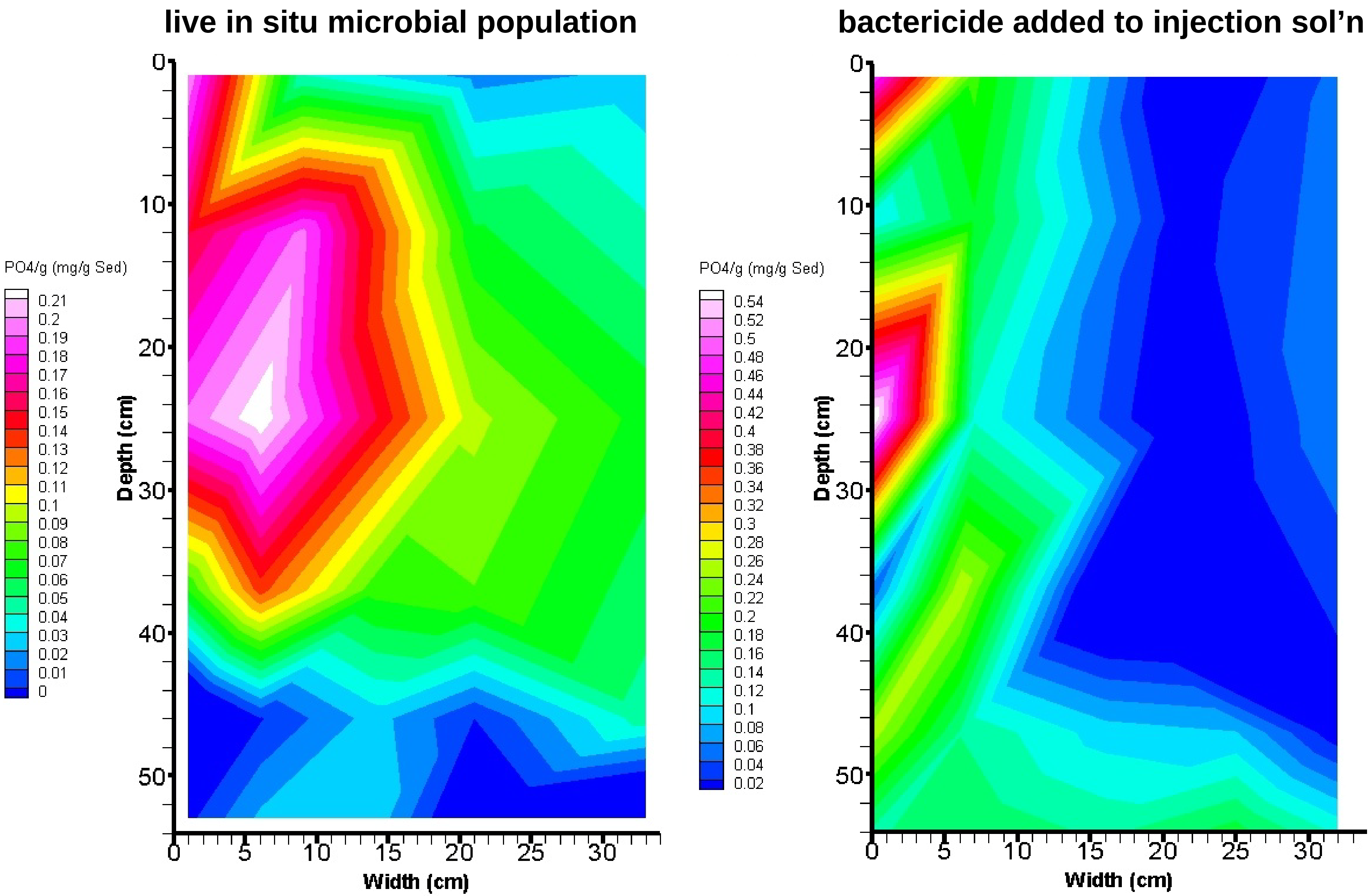
ion exchange:
- 1-10 h uptake rate
- 45% aqueous (at eq.)

incorporation:
- 10% drop in aq. = 30% incorp. (significant mass on surfaces)
- half-life 16 months (other data 5.5 months)

ion exchange:
- rapid (dispersion limited in field)
- 1% aqueous (at eq.)

incorporation:
- same half-life
- rate <u>not</u> controlled by % Sr on apatite surface (exchange is rapid)
- field data: % aq same over time; need sediment for % exch., % inc.

Need Microbes to Form Apatite



both: infiltrate 60 mM PO4, 5.0 mL/h x 64 h