The NERC **BI**ogeochemical **G**radients and **RAD**ionuclide Transport **BIGRAD** Consortium

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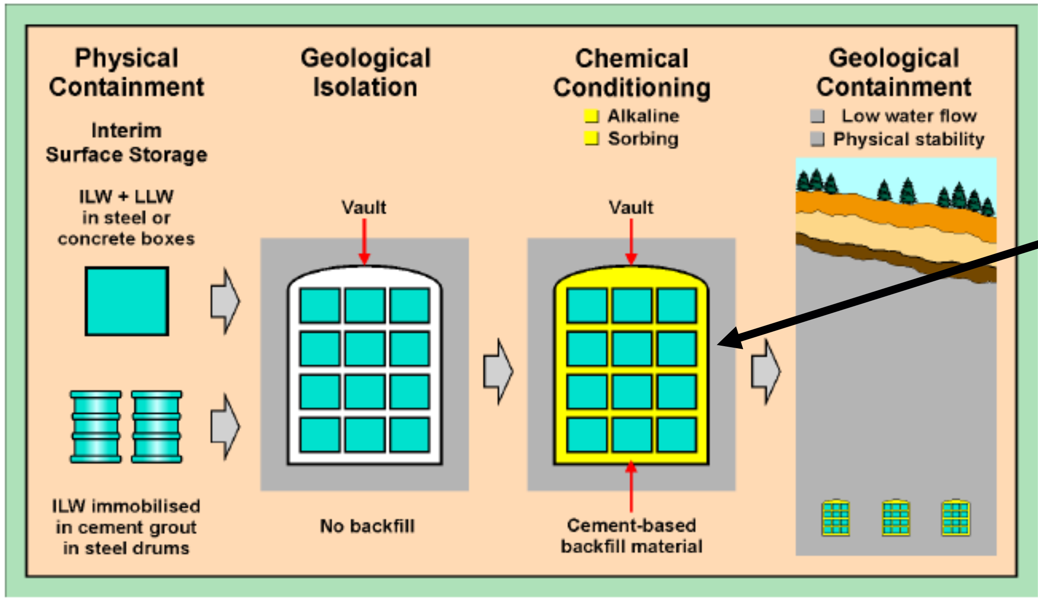
Radioactive Waste Disposal

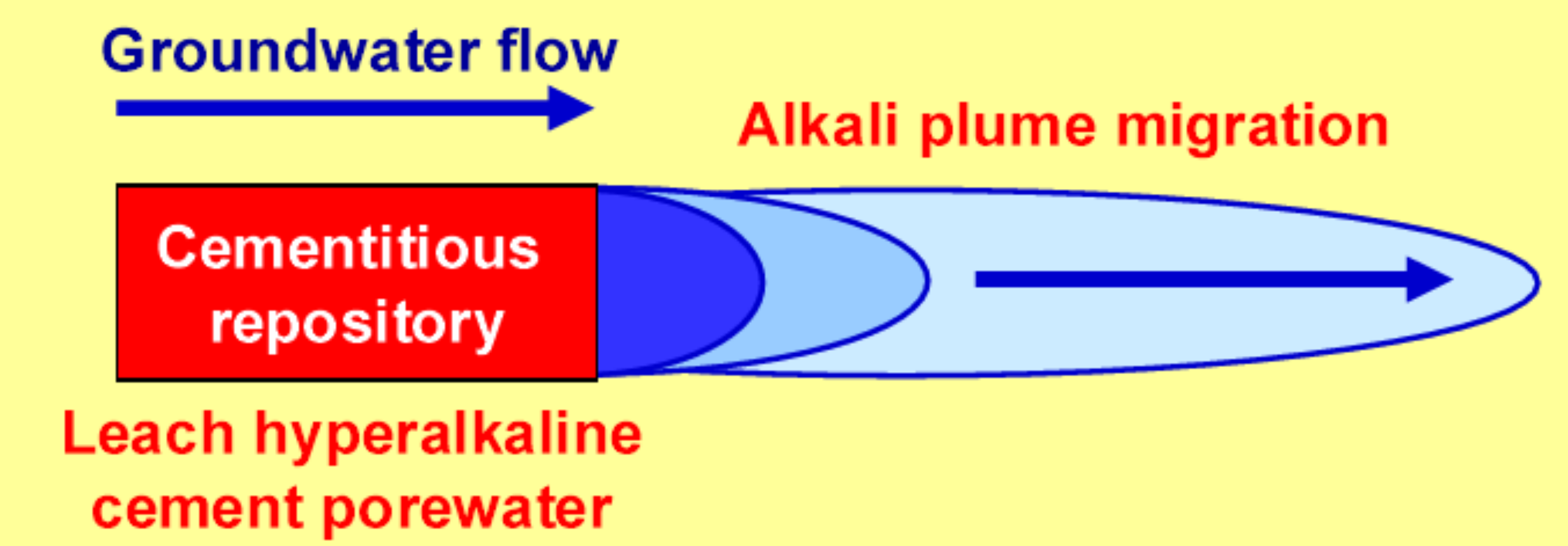
BIogeochemical Gradients and RADionuclide transport
BIGRAD
www.bigradnerc.com

TheObserver



Chemically Disturbed Zone

The Chemically Disturbed Zone



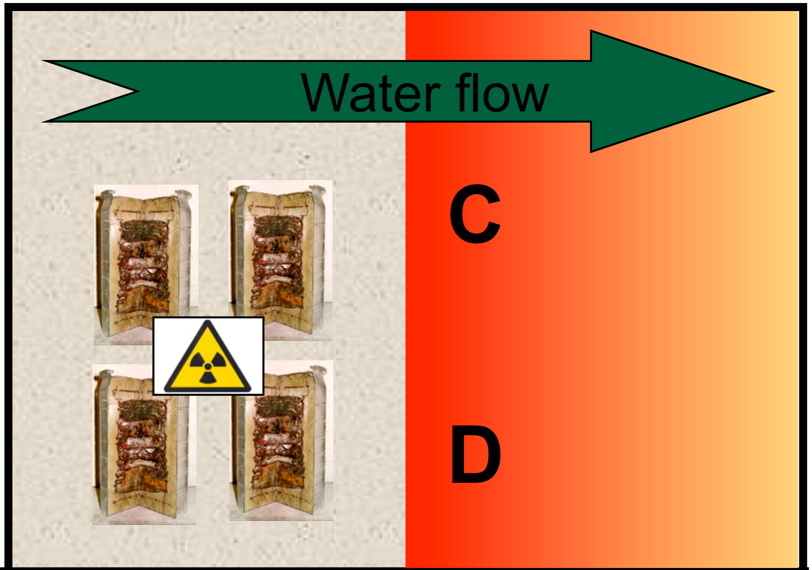
- CDZ is the dynamic interface between GDF and the natural environment
- CDZ impact on mineralogy / radionuclide transport largely unknown
- Improved knowledge → improved safety case

The CDZ is interface between waste and natural environment:

- steep, evolving biogeochemical gradients

- heterogeneous, nanometre to metre understanding required

- Tc, U, Np and Pu - evolution over 10^6 y

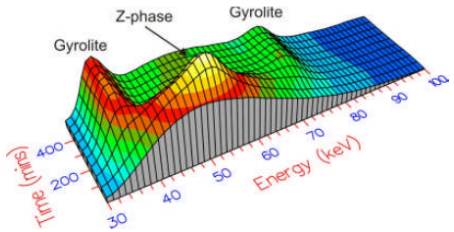


Matrix	**Cement**	**Quartz, feldspar**
pH	**Alkaline**	**6-7**
Metallic Fe	**High**	**None**
Organics	**Cellulose; fermentation products**	**Humics**
H_2	**High**	**Low**
Microbiology	**?**	**?**
Radionuclide behaviour	**✓? — — — —▶**	**?**

What is novel?

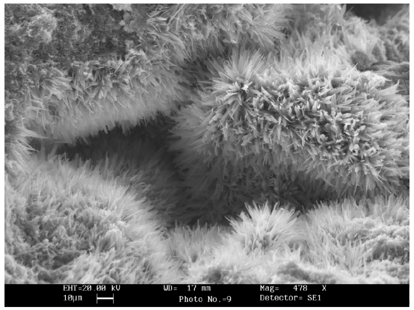
Biogeochemistry

Solution chemistry

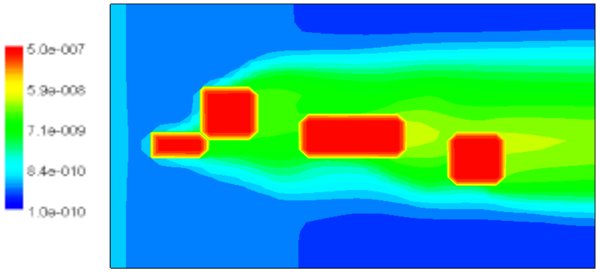


Mineral alteration

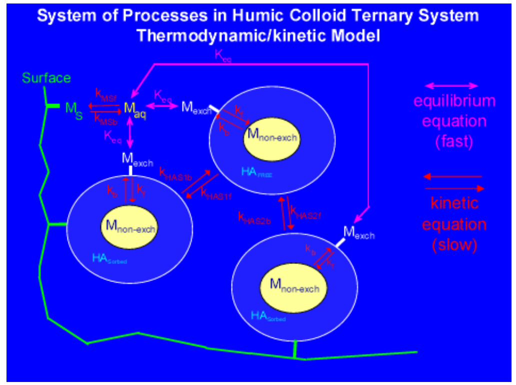
Transport



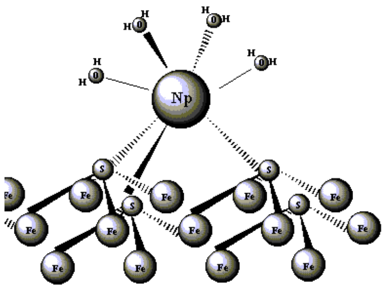
Porosity and permeability



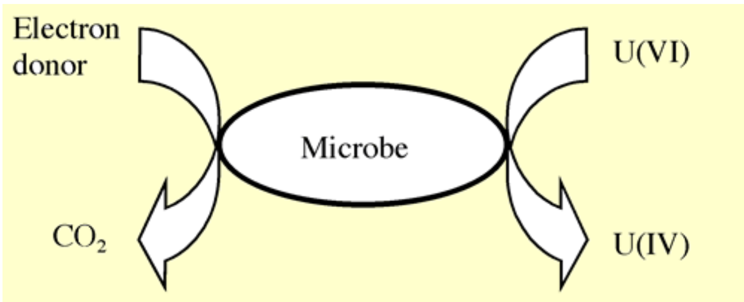
Multiscale predictive capability



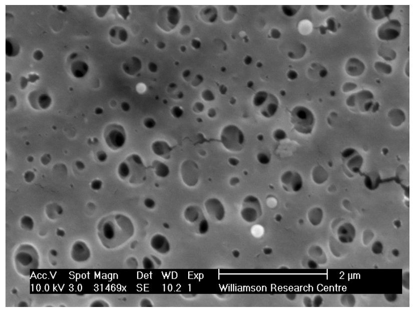
Sorption and reversibility



Radionuclide mineralogy



Radionuclide biogeochemistry



Colloidal behaviour

Scenarios / Systems

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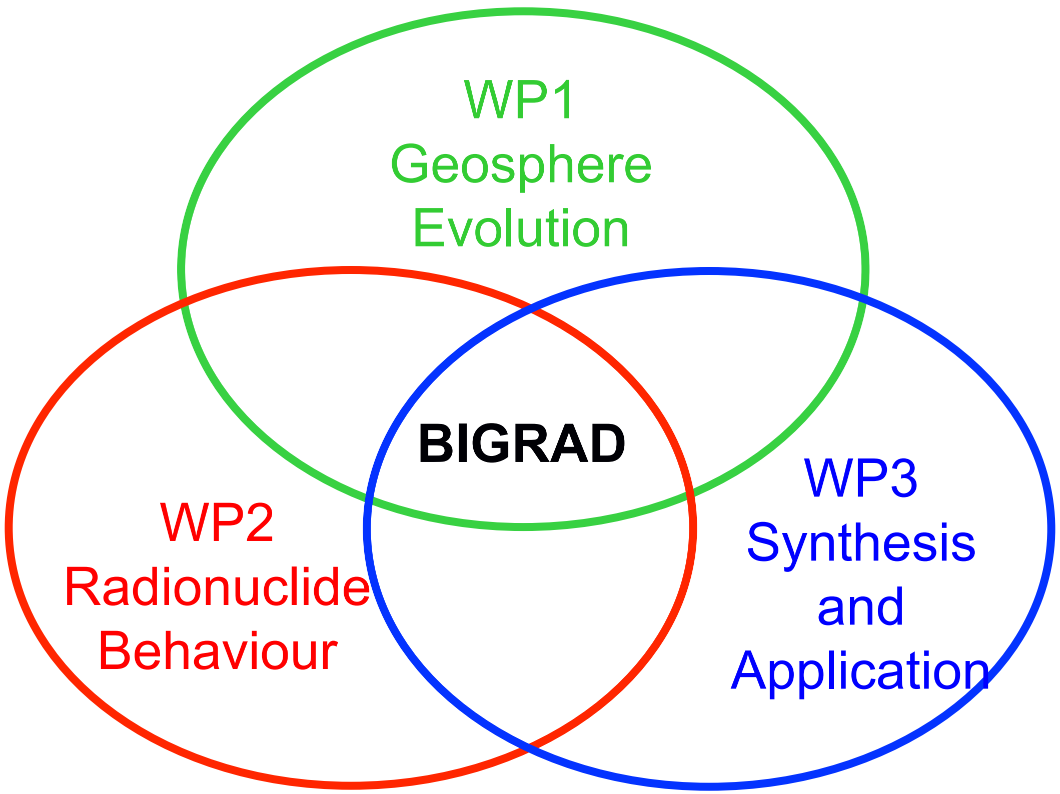
- GDF in acid / intermediate rock - likely candidate
- Minerals → mineral aggregates → model rock
- Acid / intermediate - quartz, feldspars, muscovite / chlorite / illite and iron oxides
- 4 year project, permeability, Permo-Triassic Sandstone
- Generic information relevant to GDF in a range of acid / intermediate geological settings

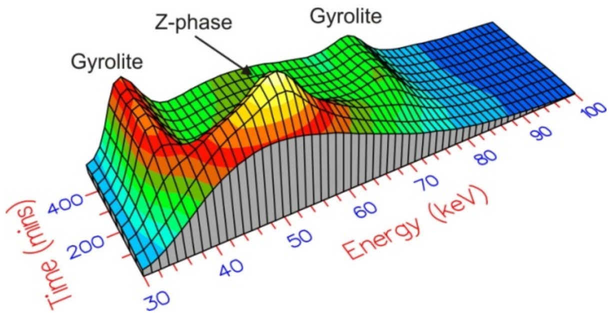
Research Hypothesis

The biogeochemical gradients that will develop across the poorly understood interface between an alkaline, deep geological disposal facility and the geosphere (the Chemically Disturbed Zone; CDZ) will be critical controls on radionuclide behaviour and transport, and thus on the safety and environmental impact of a geological disposal facility.



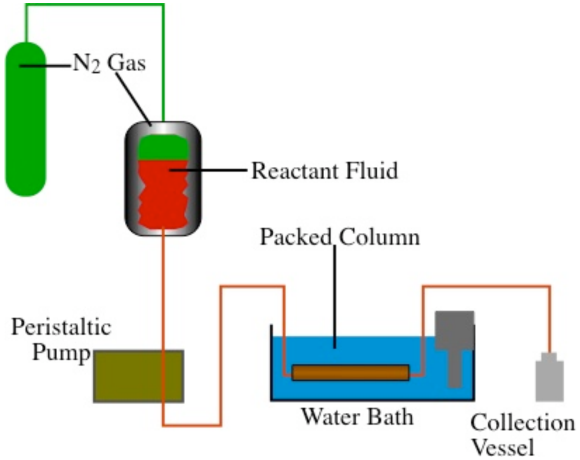
WP1 Geosphere Evolution

Mineral alteration



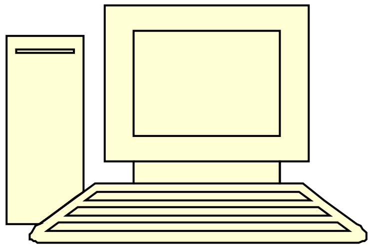
Geomicrobiology

Physical processes



WP2 Radionuclide form reaction and transport

WP3 Synthesis and application to performance assessment



Nano-scale

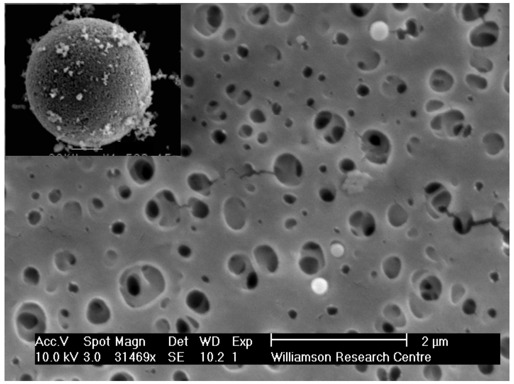
Column-scale

diamond

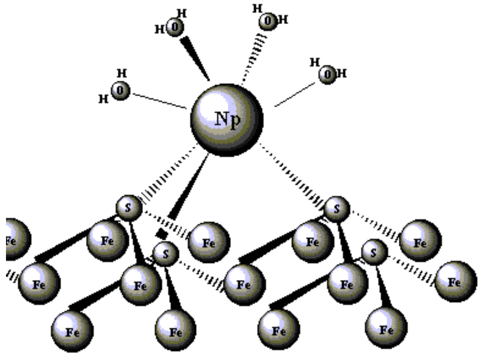
WP2 Radionuclide form, reaction and transport

Aqueous speciation

Colloids



Sorption and partitioning



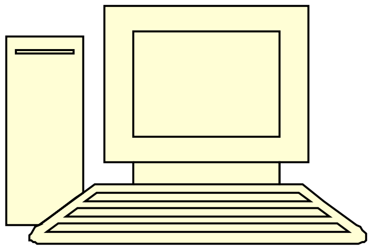
diamond

ANKA

Nano-scale

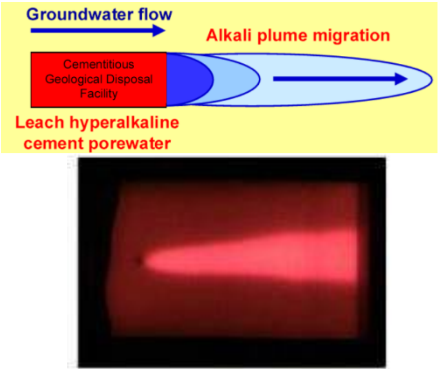
Column-scale

WP3 Synthesis and application to performance assessment



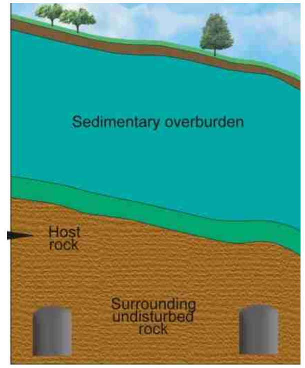
WP3 Synthesis and application to performance assessment

Conceptualisation, experimental design & bench-scale CDZ

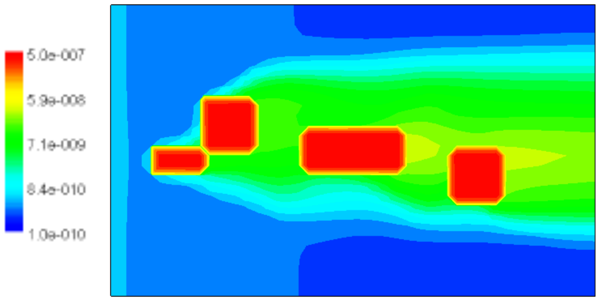


Process understanding, parameterisation with lab study data

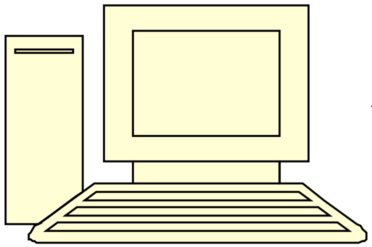
Variable GDF and geosphere settings, environmental conditions



Scenario modelling, geosphere barrier function, CDZ development

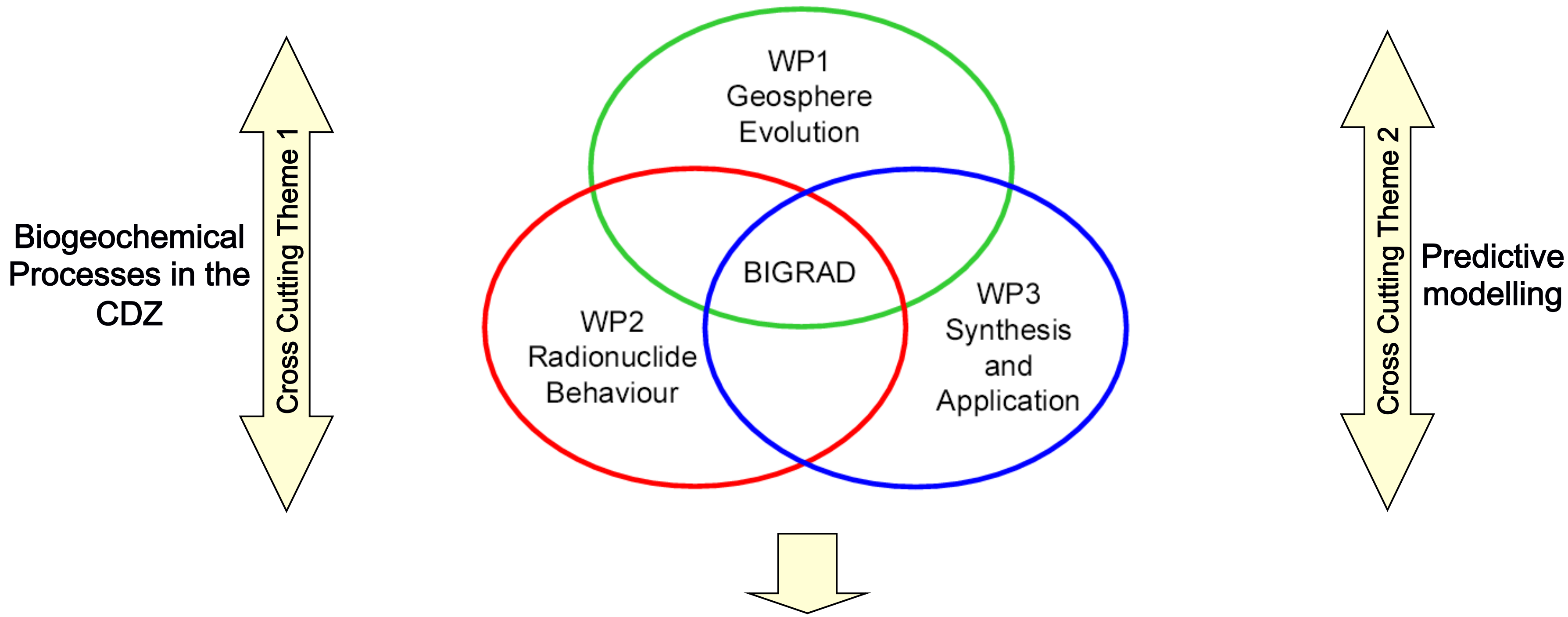


Numerical modelling tool for performance prediction



Cross Cutting Themes

- CCT 1 Biogeochemical processes / radionuclide behaviour
- CCT 2 Predictive modelling capability / radionuclide mobility



Naturally integrated WPs and CCTs require a consortium

SCIENCE

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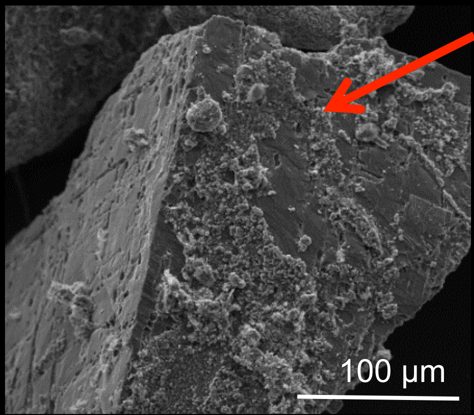
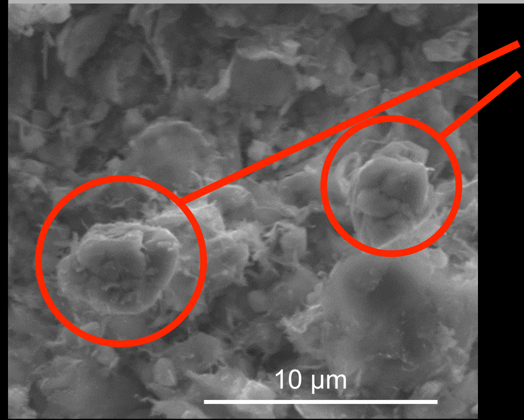
15 years BVG evolved fluid

Dolomite dissolution



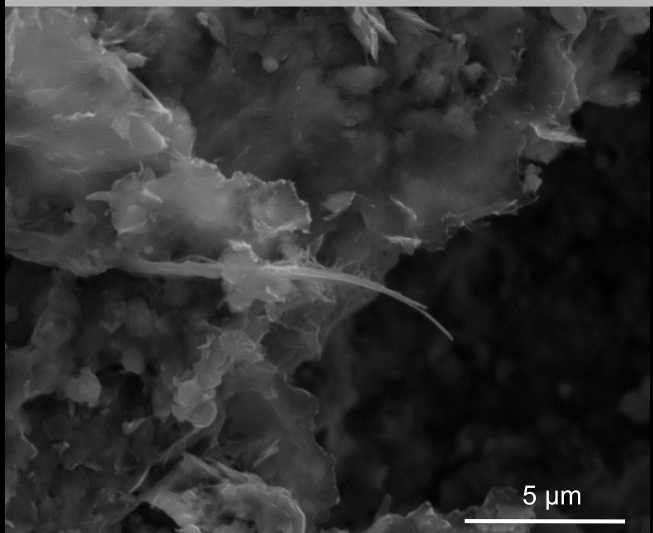
Dissolution features present on dolomite grains though less pronounced than in young fluid

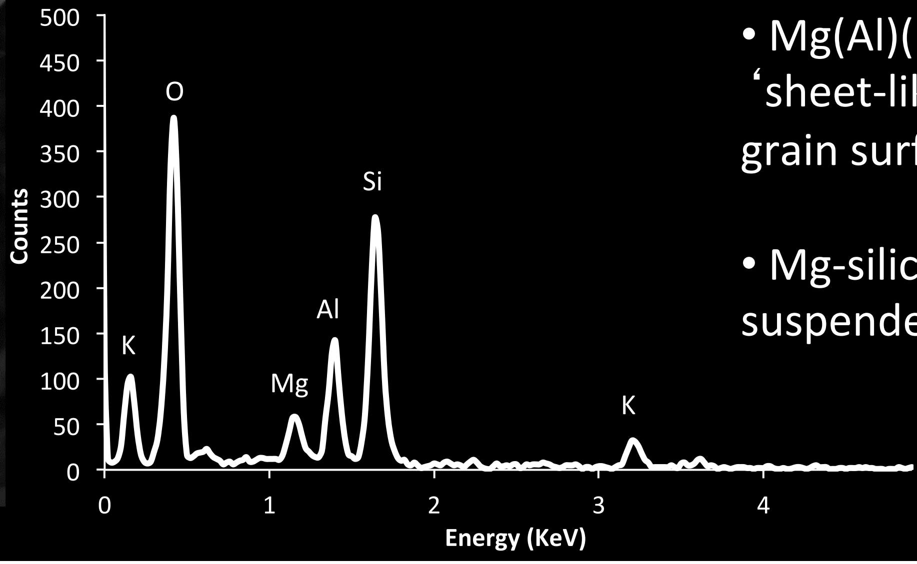
Calcite precipitation



Calcite found on grain surfaces

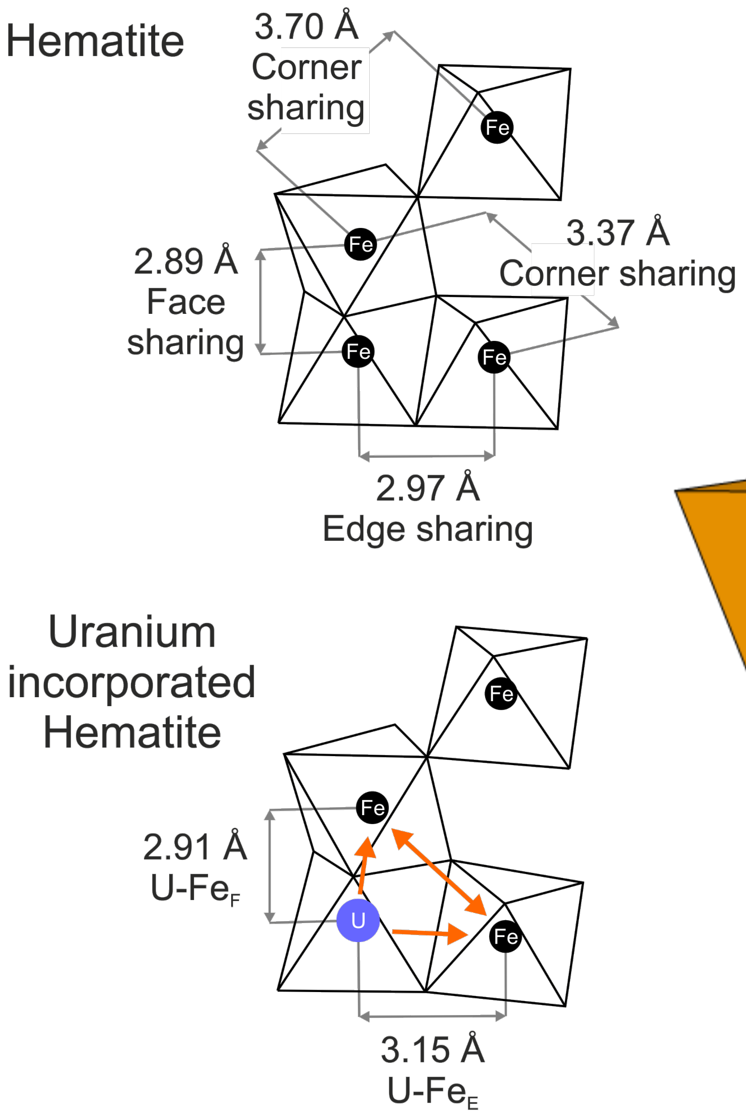
Magnesium silicates

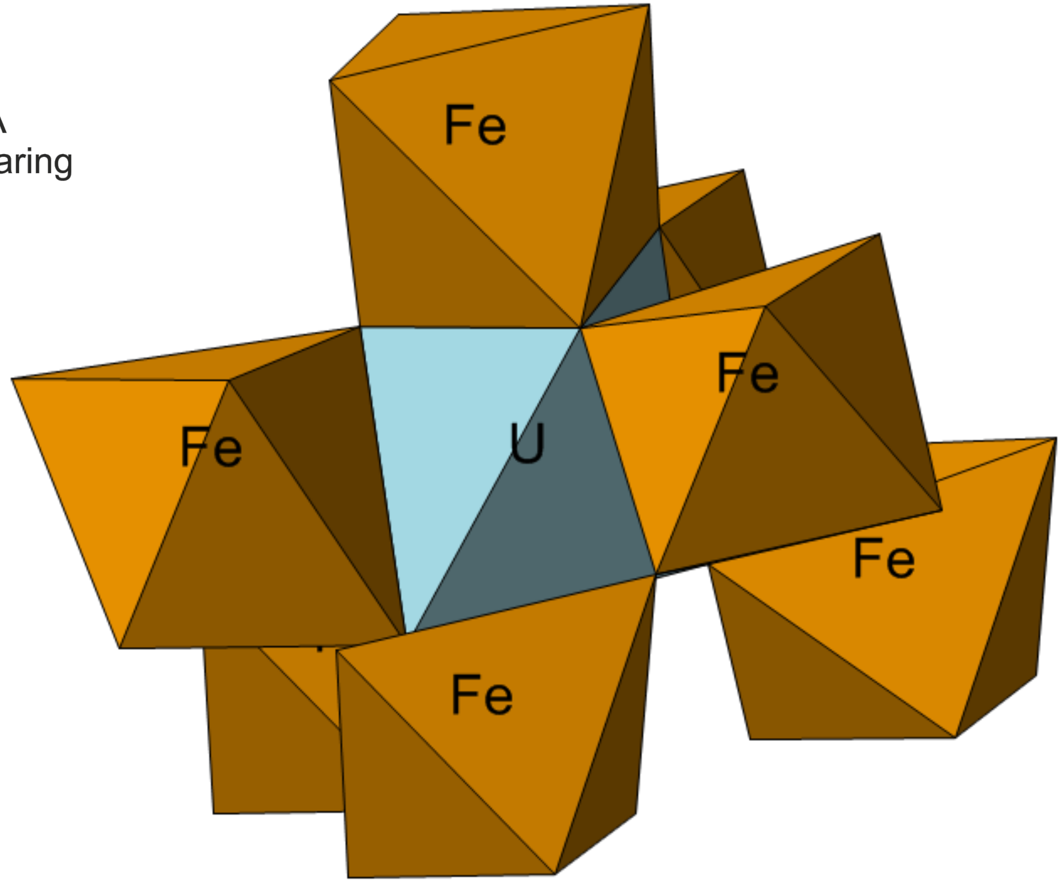




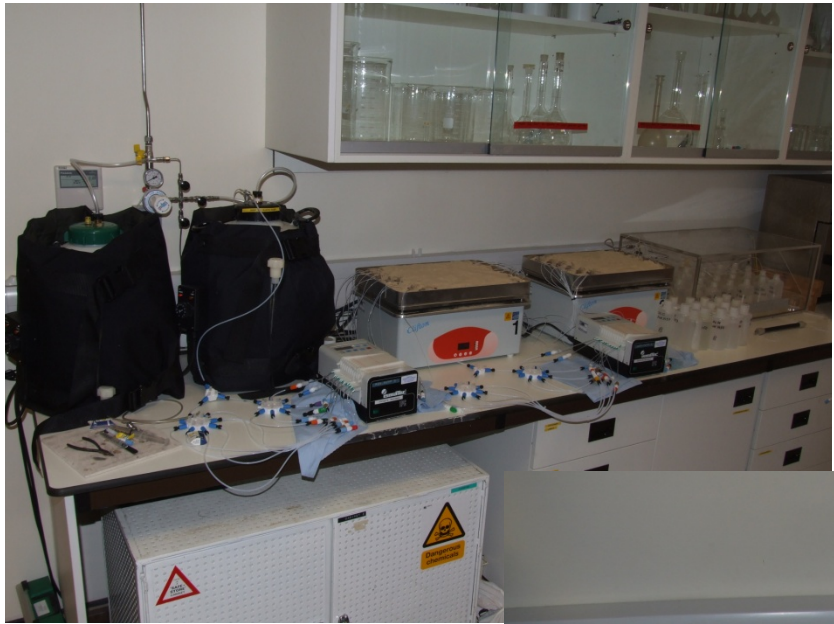
- Mg(Al)(K)-silicate 'sheet-like' phases on grain surfaces
- Mg-silicates suspended in solution

Hematite crystallisation at pH 10.5





adapted from Cornell and Schwertmann 2003




Clifton
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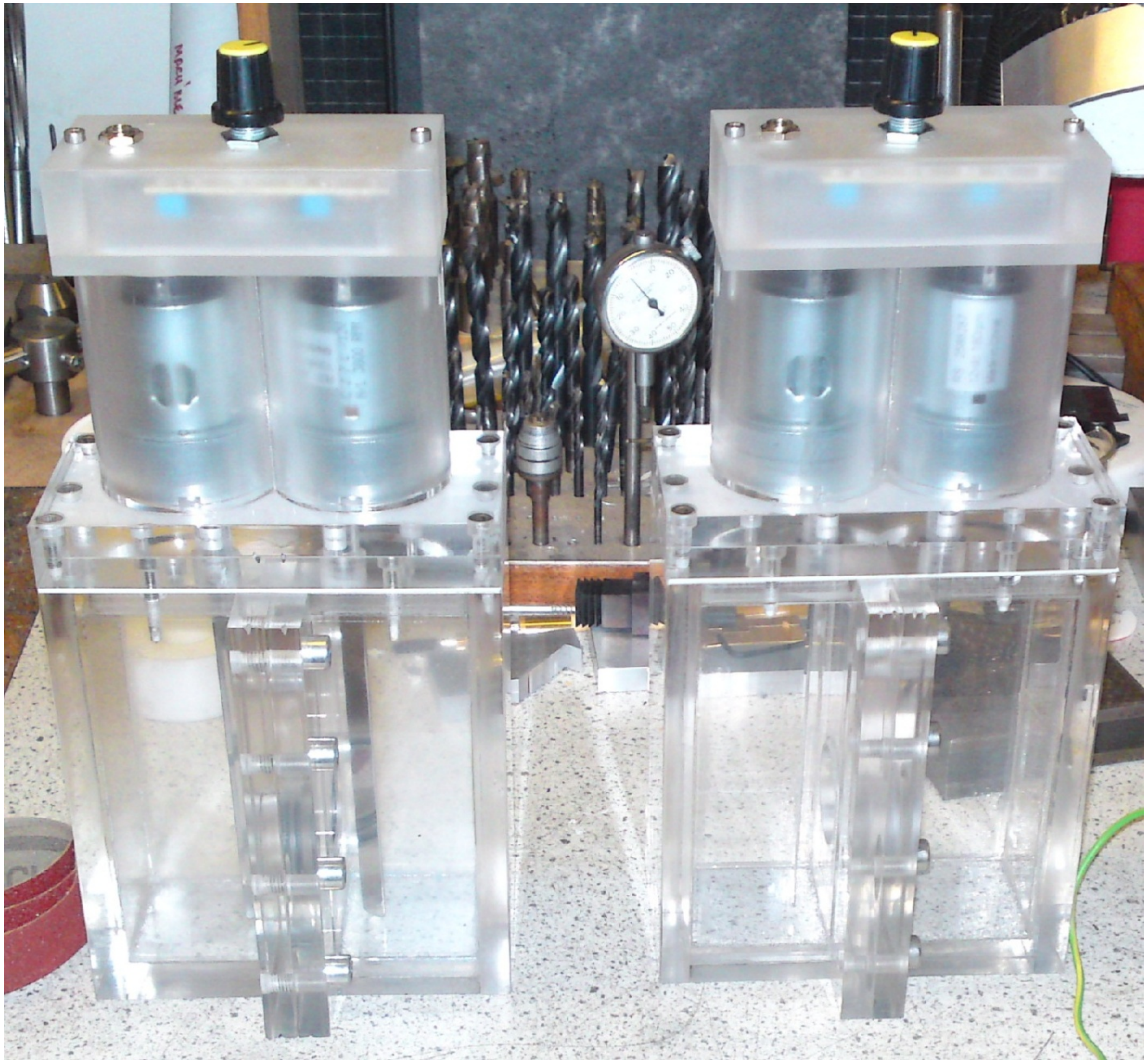
Kinetic experiments



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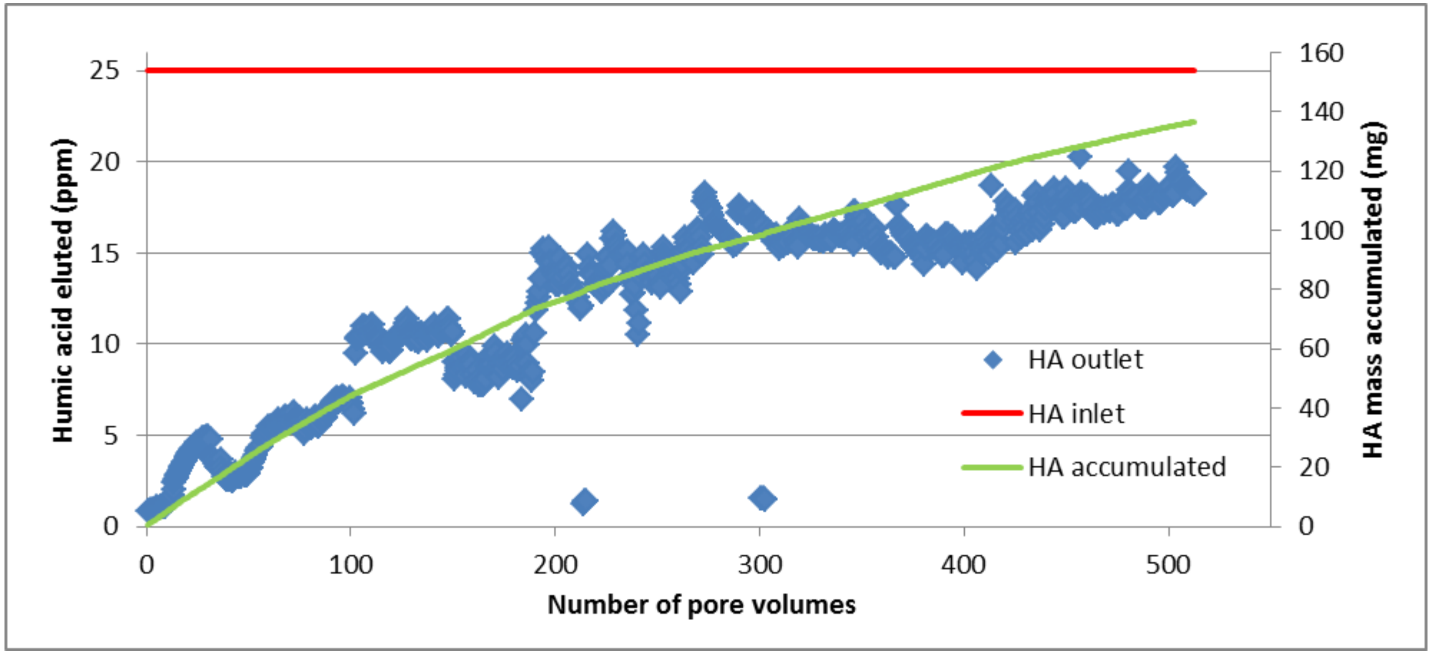
Scenario III

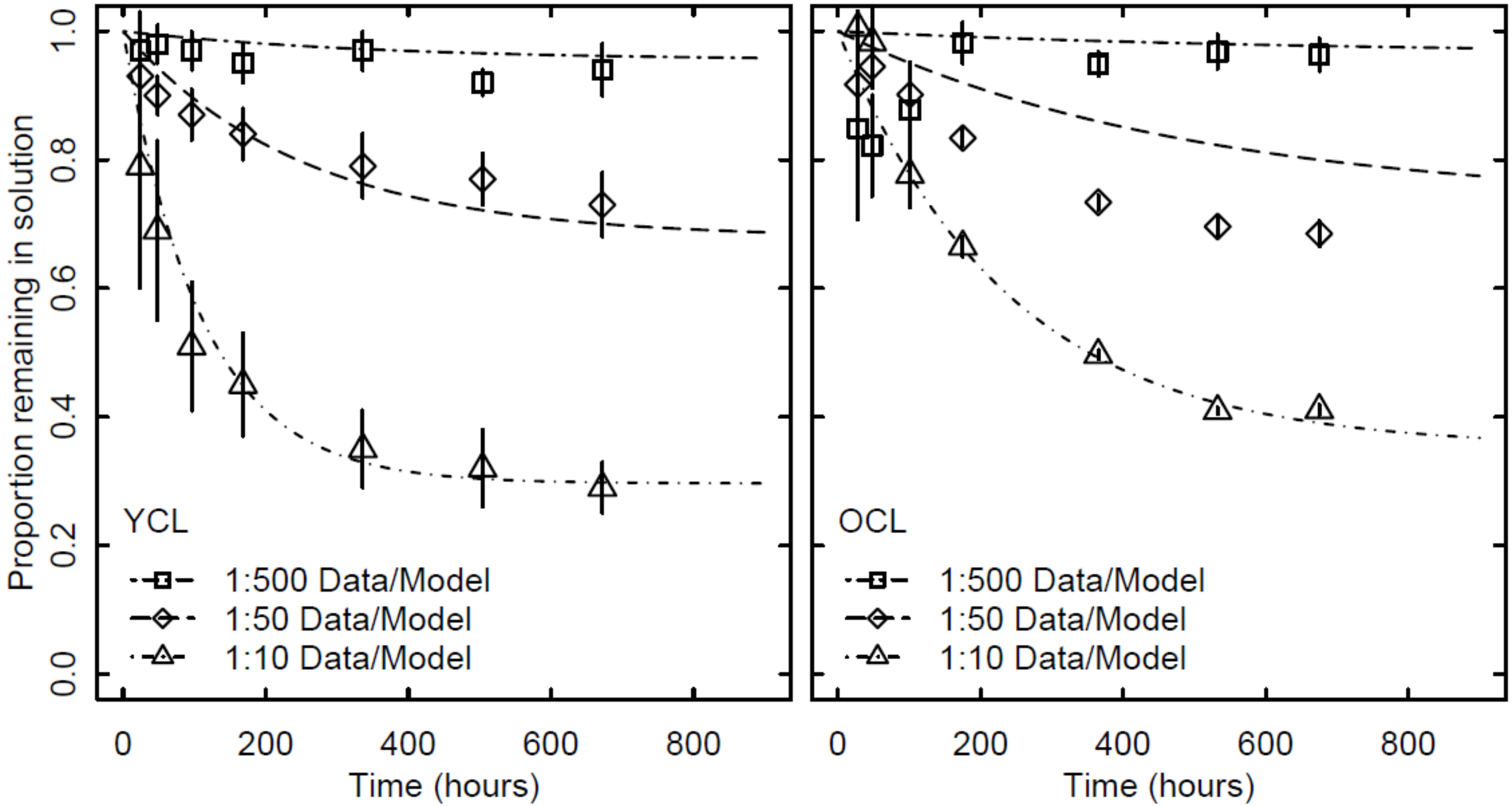
Migration of U(VI) in YCL through sandstone saturated with HA

1st stage: saturation with HA under atmospheric conditions



- Flow 0.05 mL/min
- Concentration of HA = 25 ppm, 0.1 mol/L $NaClO_4$, pH 6.5

Low level sorption



YCL rate constants of k_1 = 1.78 x 10^{-6} L mol^{-1} s^{-1} and k_2 = 7.50 x 10^{-7} s^{-1}.
OCL rate constants of k_1 = 8.20 x 10^{-7} L mol^{-1} s^{-1} and k_2 = 5.12 x 10^{-7} s^{-1}.
10 Bq ml^{-1} ^{232}U (5.27x10^{-11} M).

1 Microbial ecology of the Cyprus natural analogue

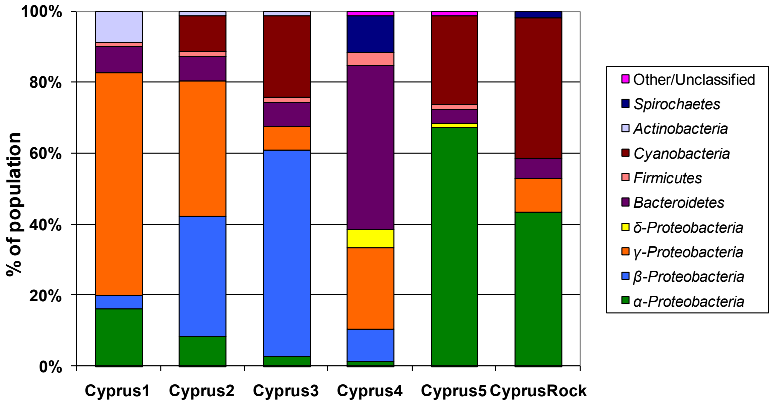
Experimental site & samples

1a 1b 1c 2 3 4a 4b 5

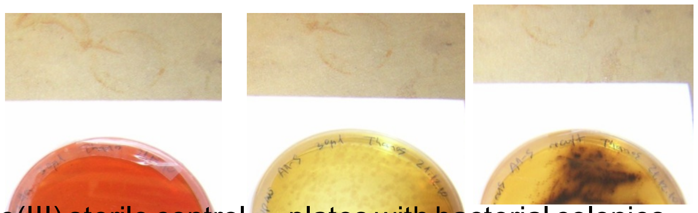
Enrichment cultures at pH 10

1a 1b 1c 2 3 4a 4b 5

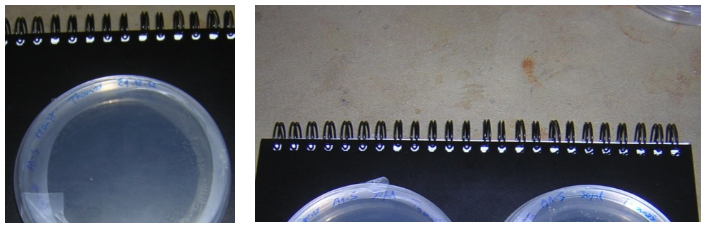
Bacterial diversity



Isolated bacteria at pH 10



Fe(III) sterile control plates with bacterial colonies



SO_4^{2-} sterile control plates with bacterial colonies

The University of Manchester

MANCHESTER 1824

Active Batch Experiments

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T0

T7

T56

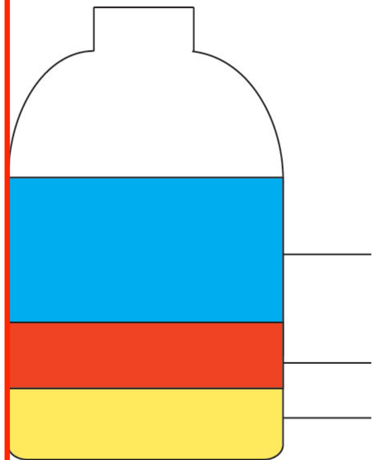
A B C D

A= Bioreducing

B=Bioreducing + HFO

C= Bioreducing + AQDS

D= Bioreducing + AQDS + HFO



100ppm U(VI)

Na Lactate (10 mM) +YE

-/+ AQDS (C+D)

+/-HFO (30 mmol L^{-1}) (B+D)

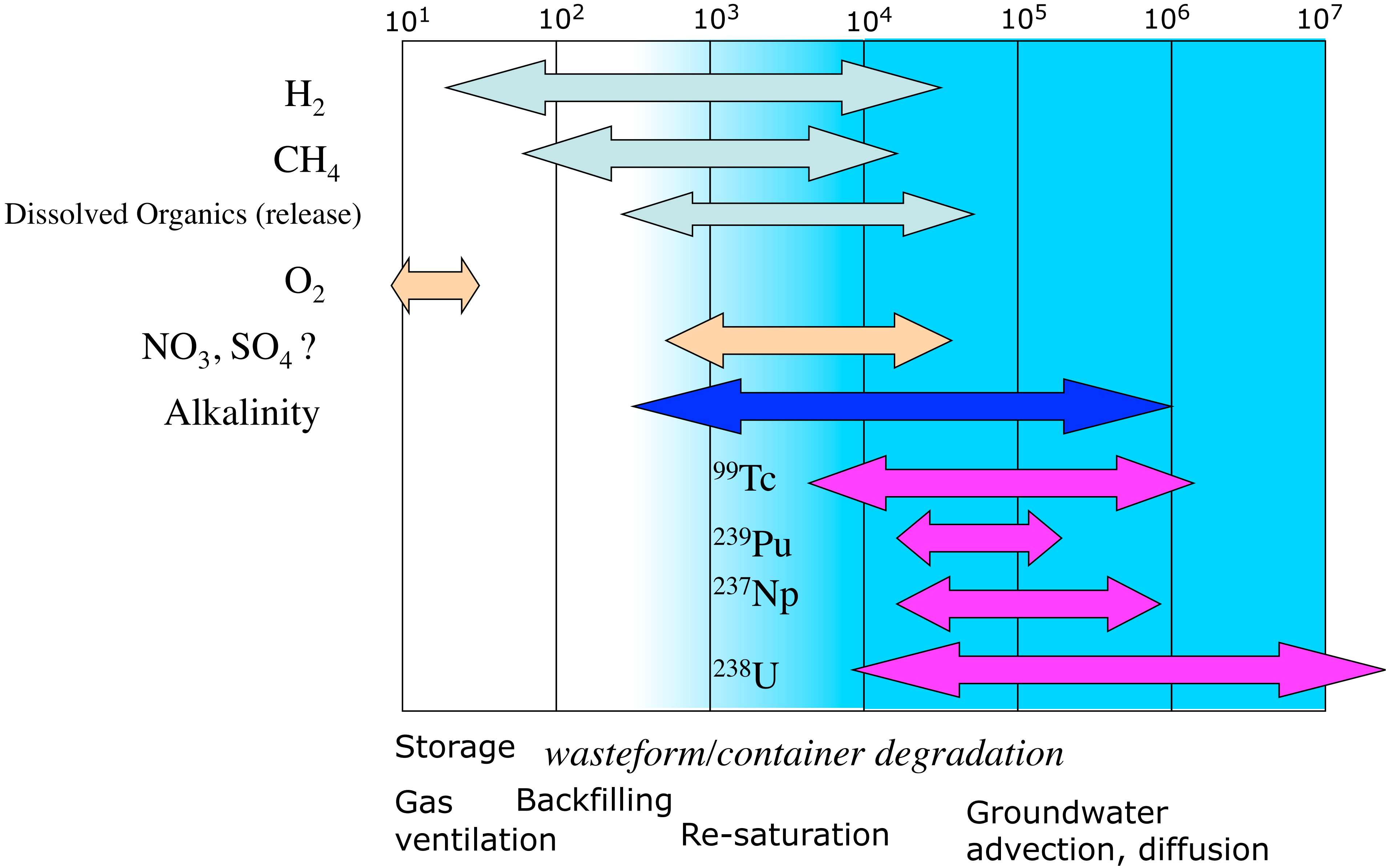
Buxton sediment

XANES on all end members

EXAFS on post reduction mineral phases

Geochemical analyses (pH, Eh, Fe(II), U(VI))

Evolution of the CDZ (time (y) after waste emplacement)



Other Developments

Env-Rad-Net (Nov 2012)

- network in the multidisciplinary field of Environmental Radioactivity
- increasing application of STFC facilities to key research challenges
- exploiting synchrotron, neutron, laser and HPC approaches
- ***Sam Shaw and Gareth Law (Manchester)***

Royal Society Industrial Fellow (NNL to 2014)

- Microbial ecology of contaminated environments
- Radiation fluxes
- Bioremediation approaches
- ***Jon Lloyd (Manchester)***