MANCHESTER 1824

The University of Manchester

Centre for Radiochemistry Research

BELBaR

Kick-off Meeting

Contribution of
The University of Manchester,
Centre for Radiochemistry Research

THE CENTRE FOR RADIOCHEMISTRY RESEARCH

Established 1999 as a partnership with nuclear industry

Facilities – Unique in the UK university sector to handle high specific activity isotopes, including Np, Pu.

Current research programmes:
Environmental Radiochemistry;
Actinide Coordination Chemistry;
Chemistry of Waste Disposal;

EPSRC Nuclear FiRST DTC.

Sponsors: EPSRC Engineering and Physical Sciences Research Council
NATURAL ENVIRONMENT RESEARCH COUNCIL
Home Office BUILDING A SAFE, JUST AND TOLERANT SOCIETY
NDA

Manchester Work

Aim: To study radionuclide interactions with Bentonite colloids and in particular (pseudo-) irreversibility in binary and ternary systems.

Our work plan is based upon our previous experience with radionuclide/humic interactions, and the prediction of radionuclide partition in mineral-humic acid ternary systems. Also on our work on radionuclide-inorganic colloid interactions in UK spent fuel storage ponds.

WP1

Concentrating on measurement and effect of irreversibility of radionuclide binding to colloids .

Manchester WP 3
BELBaR Materials

Bentonite (colloid):

We plan to develop our methods using Ca-Bentonite and (Wyoming) Na-Bentonite. When they are refined, we have a small sample of FEBEX Bentonite (thanks to CIEMAT!).

'Rock':

Single/simple phases (silica, quartz sand, etc.); Granite; Sherwood sandstone (study material for large UK CDZ project (BIGRAD); bentonite.

Radionuclides:

^{152}Eu for method development, then Am, Pu and U (^{232}U and $^{235/238}$U to allow low concentrations, exchange experiments).

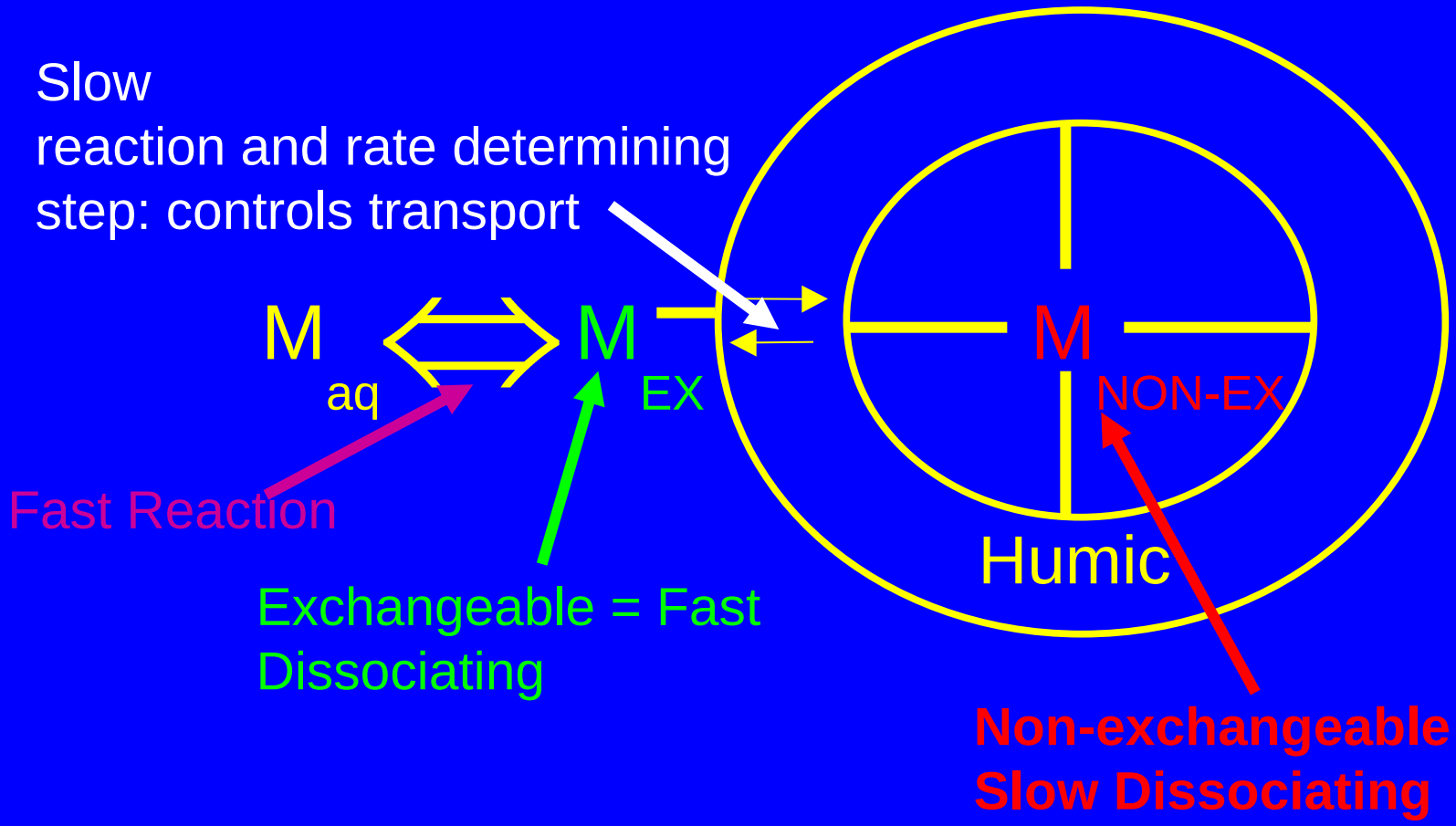
These are proposals, but we are flexible

For Humics.....

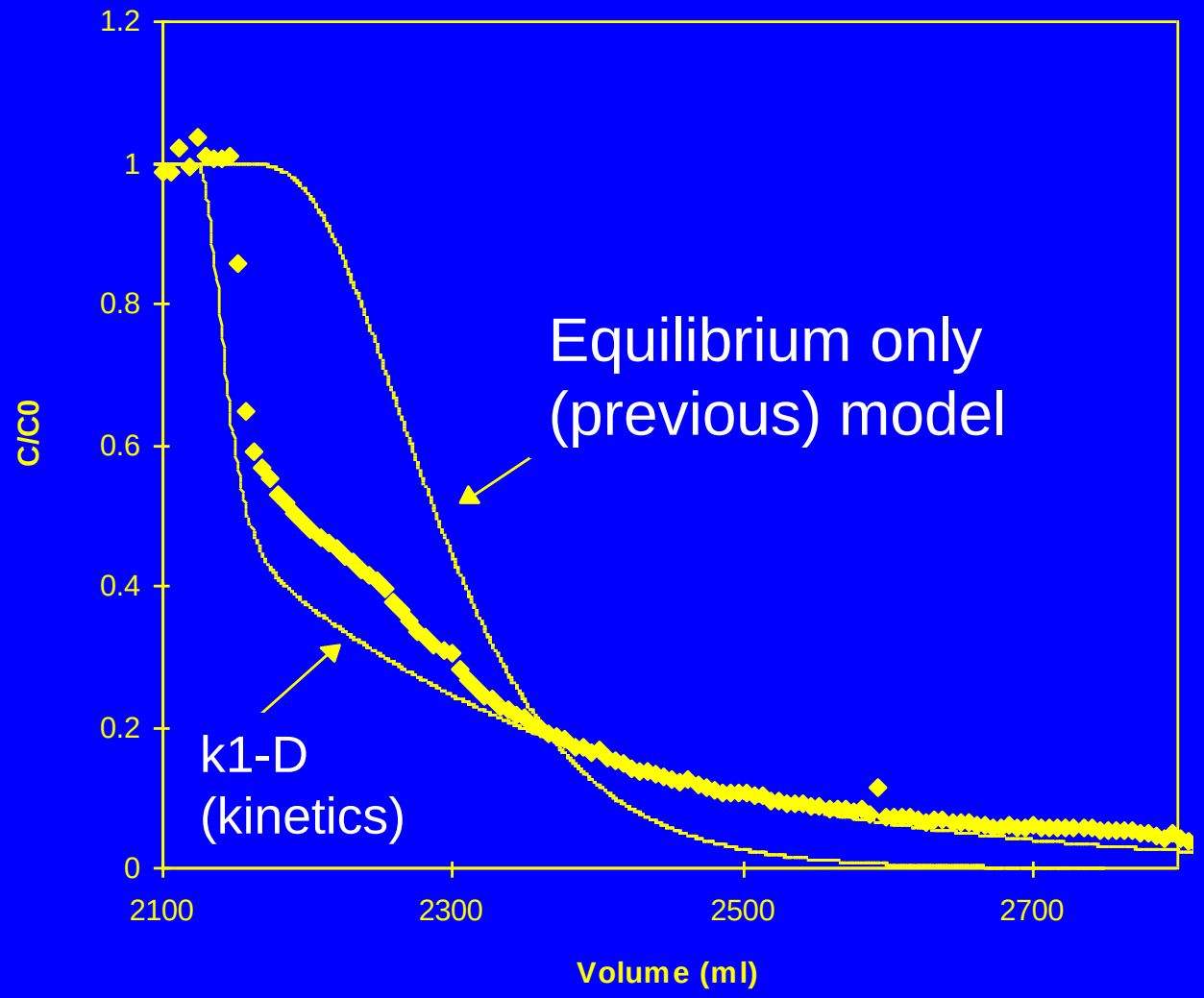
Humic complexes do not dissociate instantaneously

Two binding modes: exchangeable and non-exchangeable

Slow
reaction and rate determining
step: controls transport

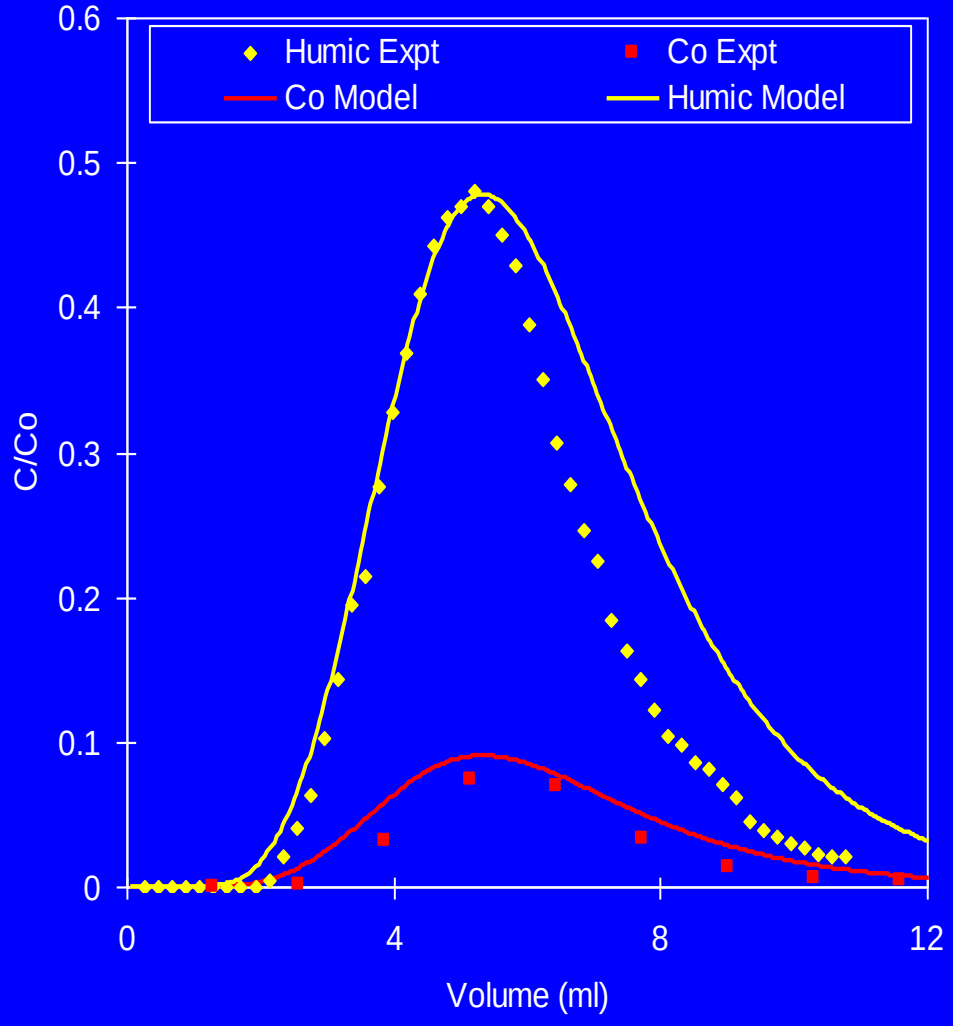


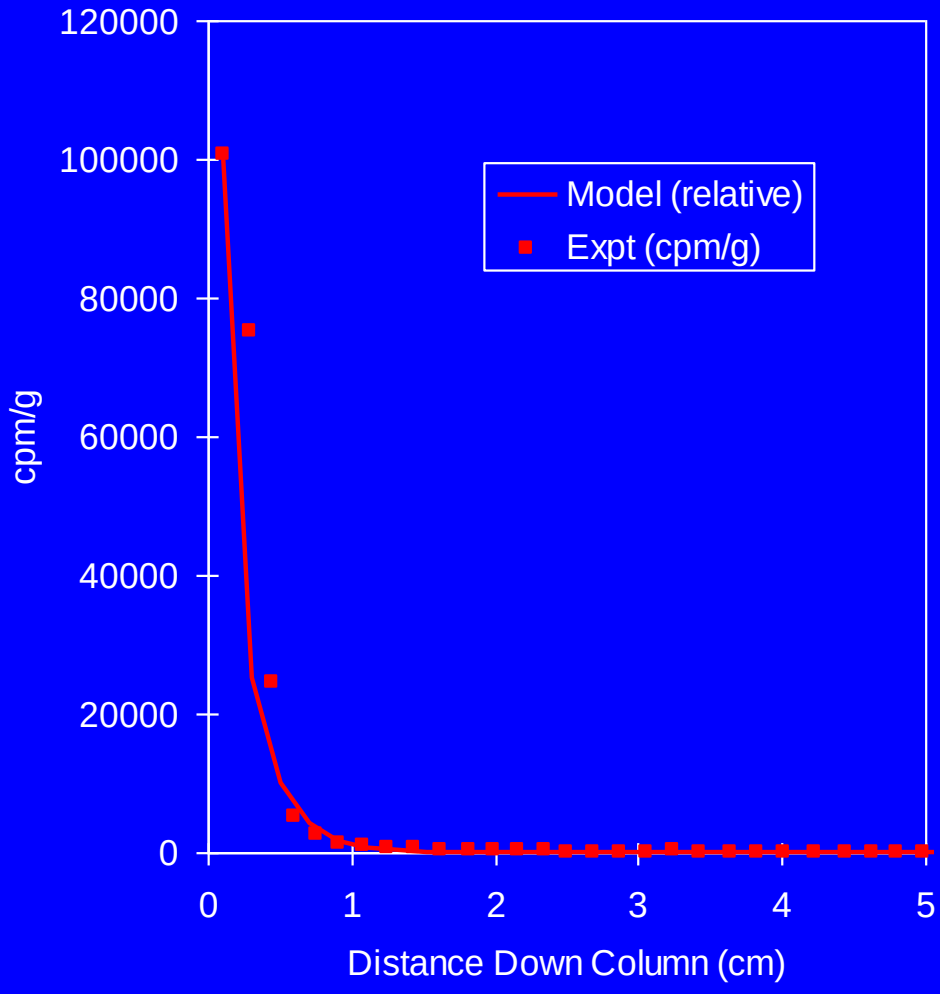
Effect of Including Slow Dissociation Kinetics in Transport Modelling



Experimental Data from University of Loughborough

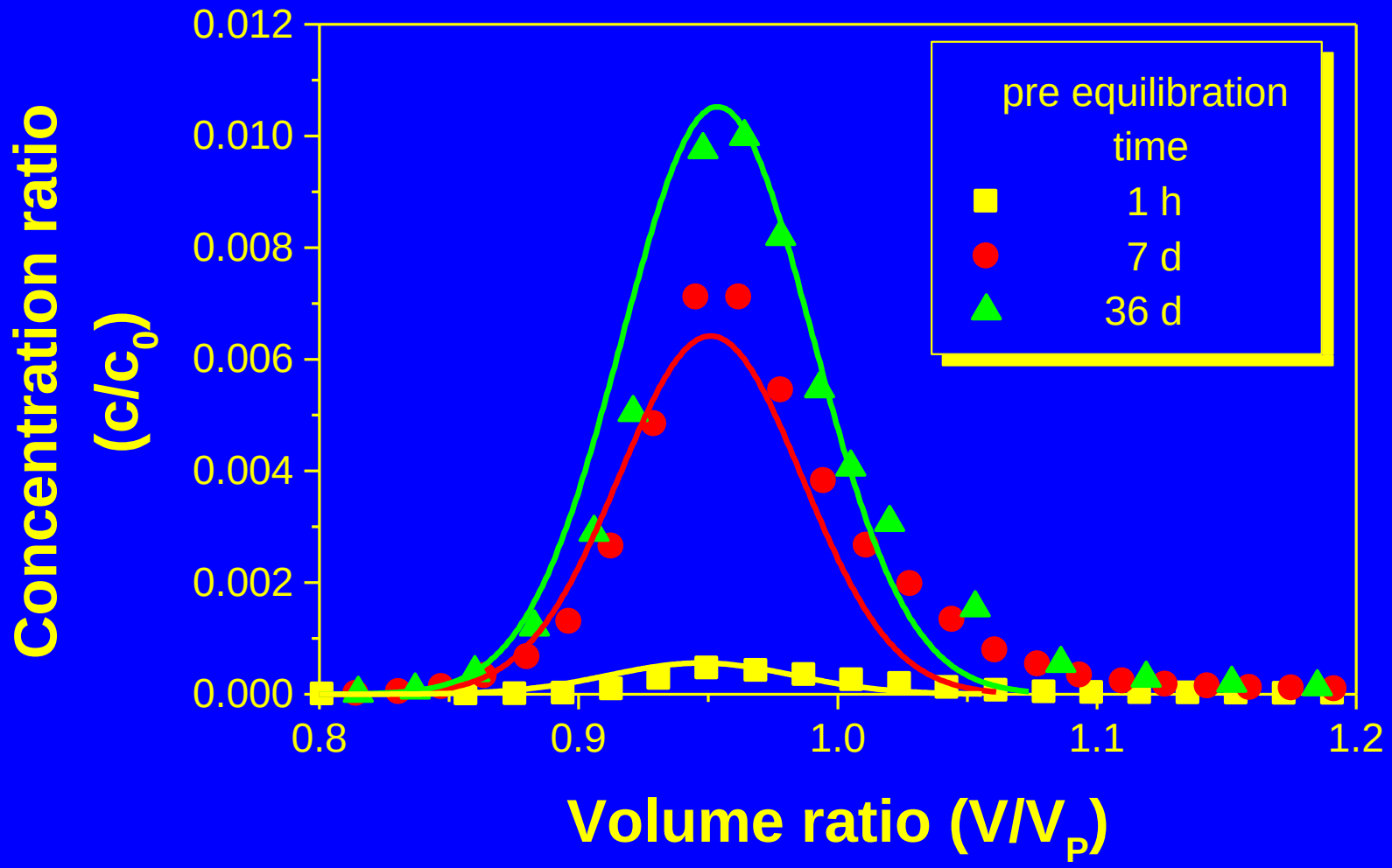
Humic and Co^{2+} Column Experiments





Experimental Data from BGS

Am^{3+} breakthrough curves and model fits
(GoHy-2227 Groundwater)

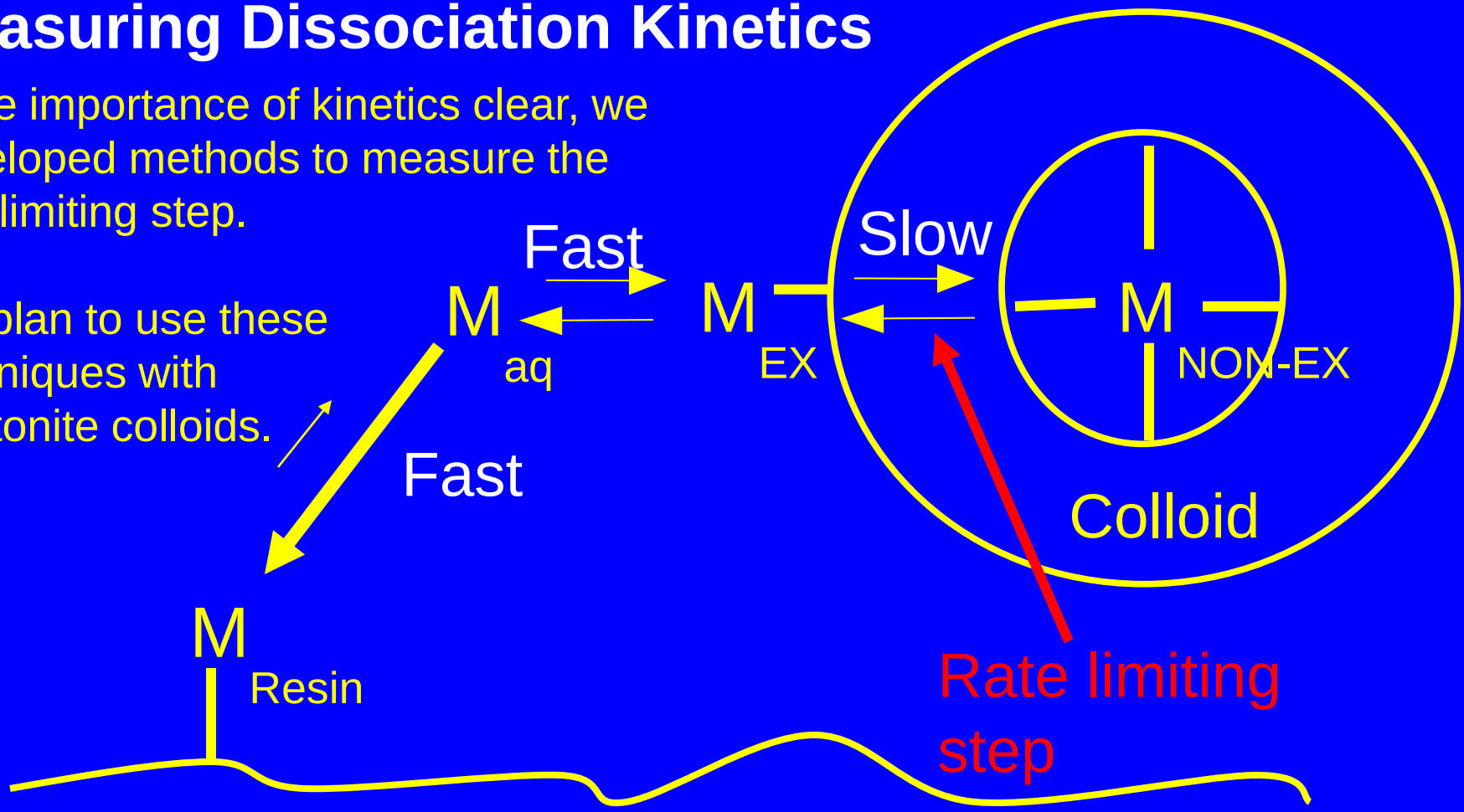


Experimental Data from KIT-INE

Measuring Dissociation Kinetics

Once importance of kinetics clear, we developed methods to measure the rate limiting step.

We plan to use these techniques with Bentonite colloids.



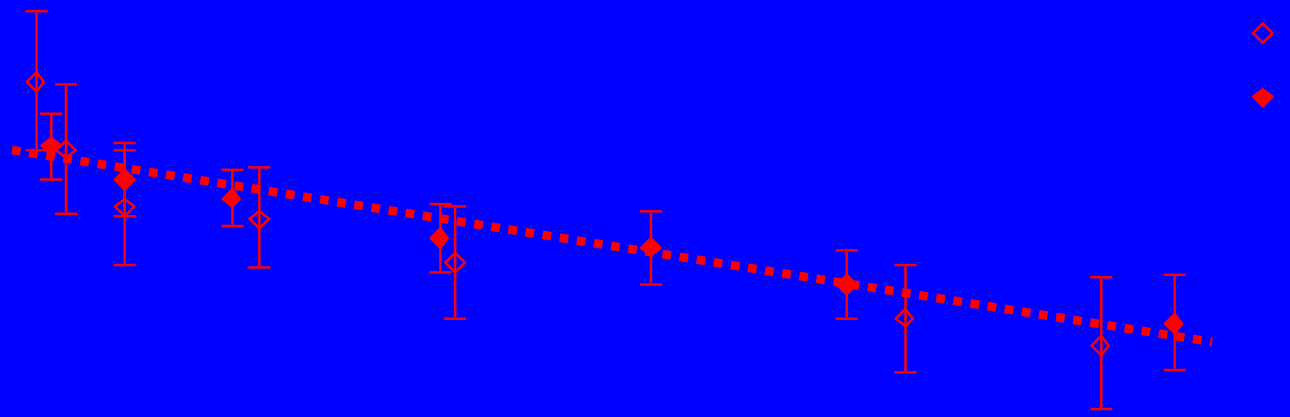
Exchange Resin (Excess Strong Sink)

Radionuclide remaining in solution = non-exchangeable.

Plot as ln [M-HA] vs time → rate constant

Example of Data from Resin Technique

^{241}Am Dissociation Data from humic sample from Sellafield

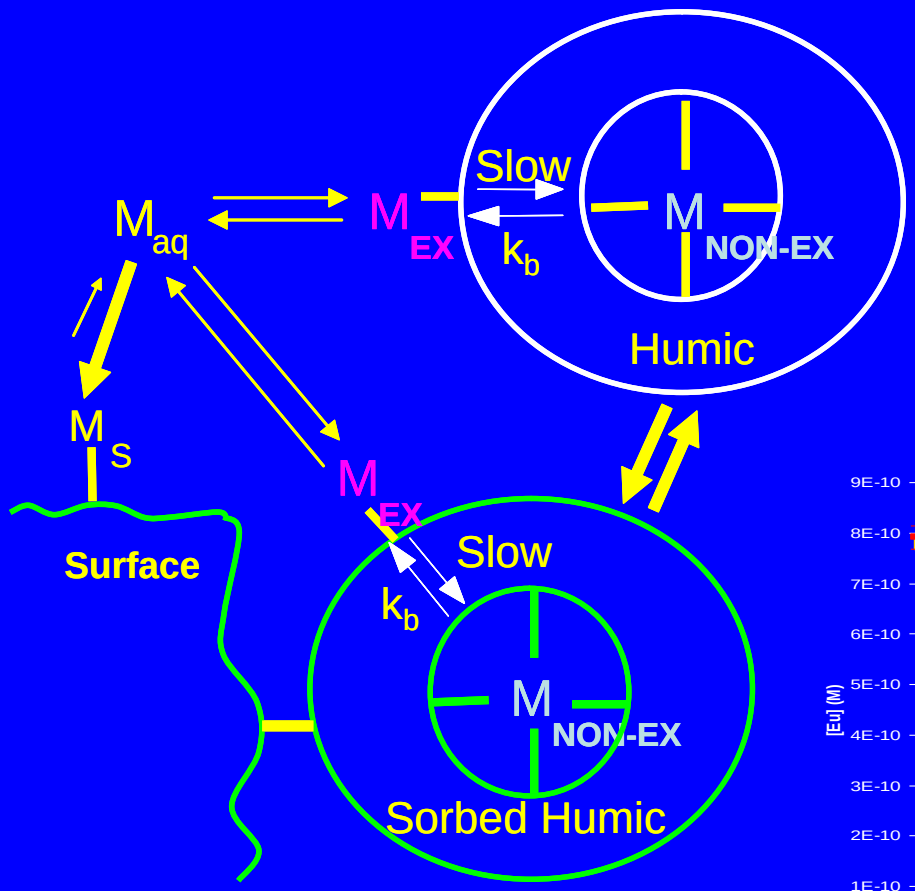


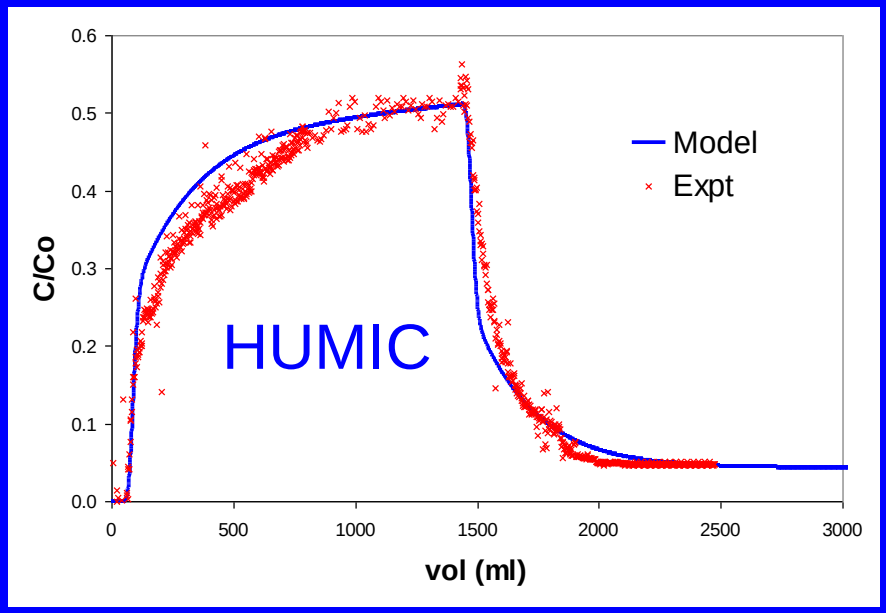
Experiment	Rate constant (s^{-1}) (2σ error)	$(C/Co)_{t=0}$ (%) (2σ error)
1	6.3×10^{-8} (19 %)	26 (3)
2	5.9×10^{-8} (18 %)	28 (2)

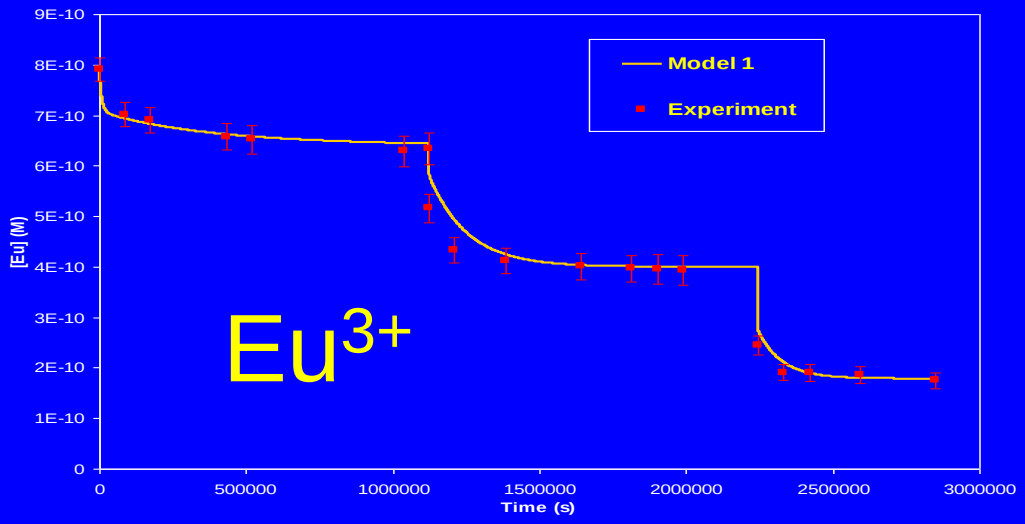
Previous (humic) Ternary System Work

Developed model of simple ion

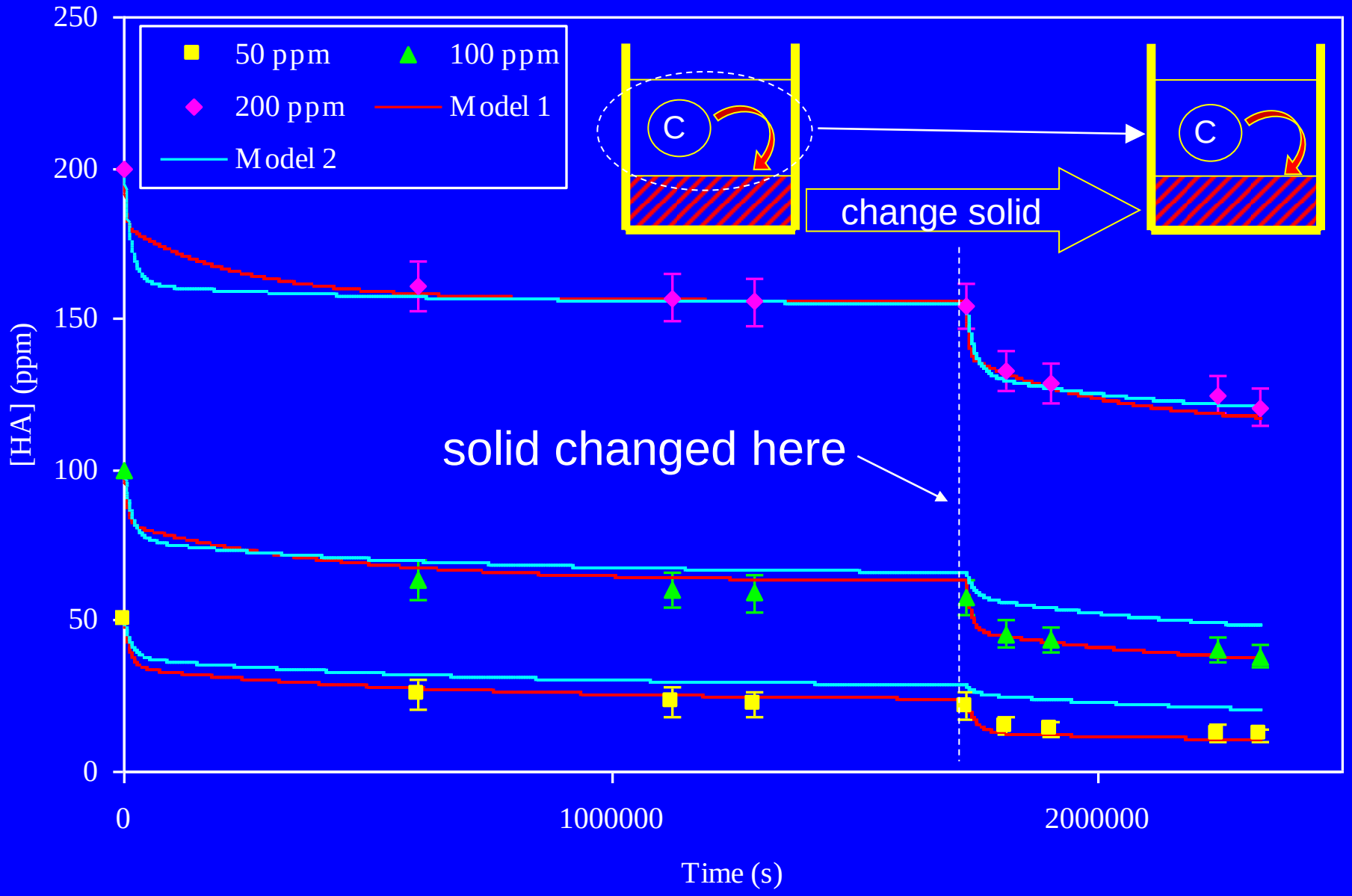
(e.g. Eu^{3+}) behaviour.







Sequential Sorption Experiments



These were very useful in defining mechanisms in humic ternary systems

Our BELBaR Work (Our proposals)

Measure extent of reversibility of RN interactions (dissociation rate constants). Effect of:

- Equilibration time;
- Ionic Strength/pH (within range of stability of bentonite colloids);
- RN and oxidation state;
- Colloid size (prepared by ultra-filtration).

Full Ternary System Experiments, as well as factors above:

- Order of addition
- Sequential sorption

'Memory effects': Equilibrate under one set of conditions (e.g. I, pH, then change and measure effect.

Model development.

Contact Us

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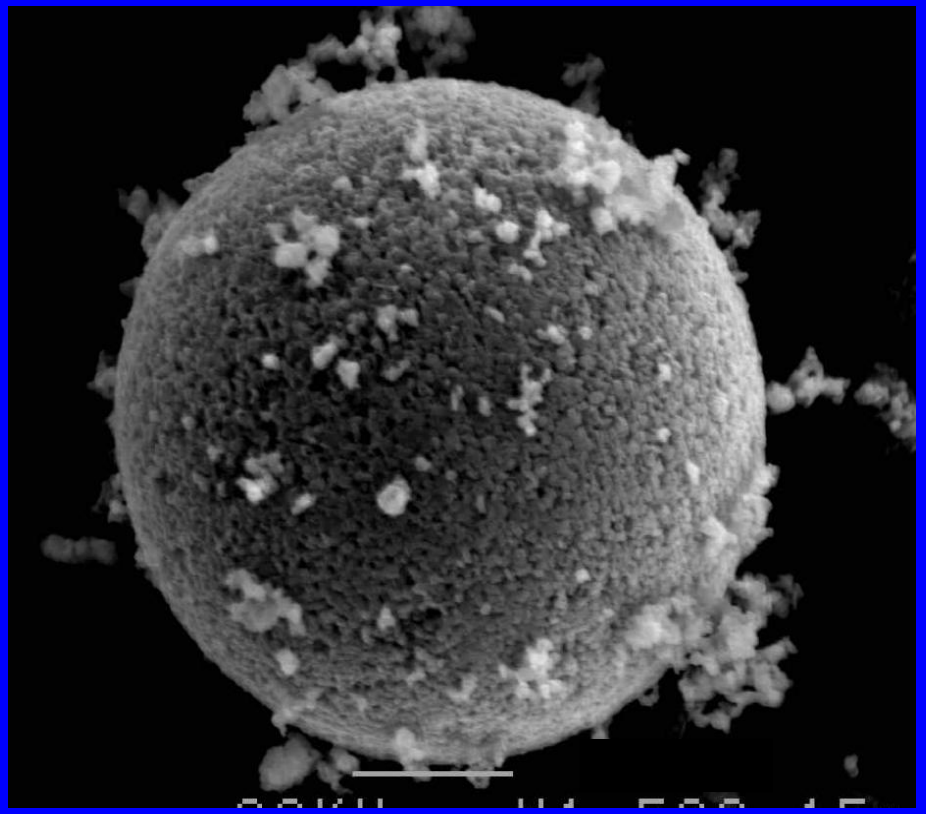

Thorium (IV) Sorption in Quartz Sand/Humic Ternary System