Business from technology

Matrix diffusion and natural analogues

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What is a natural analogue?

- Natural system that somehow resembles processes and materials relevant in nuclear waste disposal
    - Chemical analogue
    - Material analogue
    - Process analogue
- The goodness of an analogue argument
    - Depends on the similarity between the analogue system and disposal system

Natural analogue as a natural ”experiment”

- Experimental conditions cannot be controlled
  - The conditions are difficult to define in detail
- Duration of experiment is often long-term and thus relevant to disposal in contrast to lab or field experiments
- Scale of experiment often relevant to disposal in contrast to lab experiments
- Experimental conditions are in situ (from analogue point of view) in contrast to lab or field experiments
  - Processes have occurred before any man-made disturbance
- Quantitative interpretation of a natural experiment is demanding
  - Conclusions are often site specific

Why should one study natural analogues?

- Test conceptual models
  - Coupled processes in situ
  - Glacial cycles
- Test the goodness of different conceptual models in interpreting the observations

Background

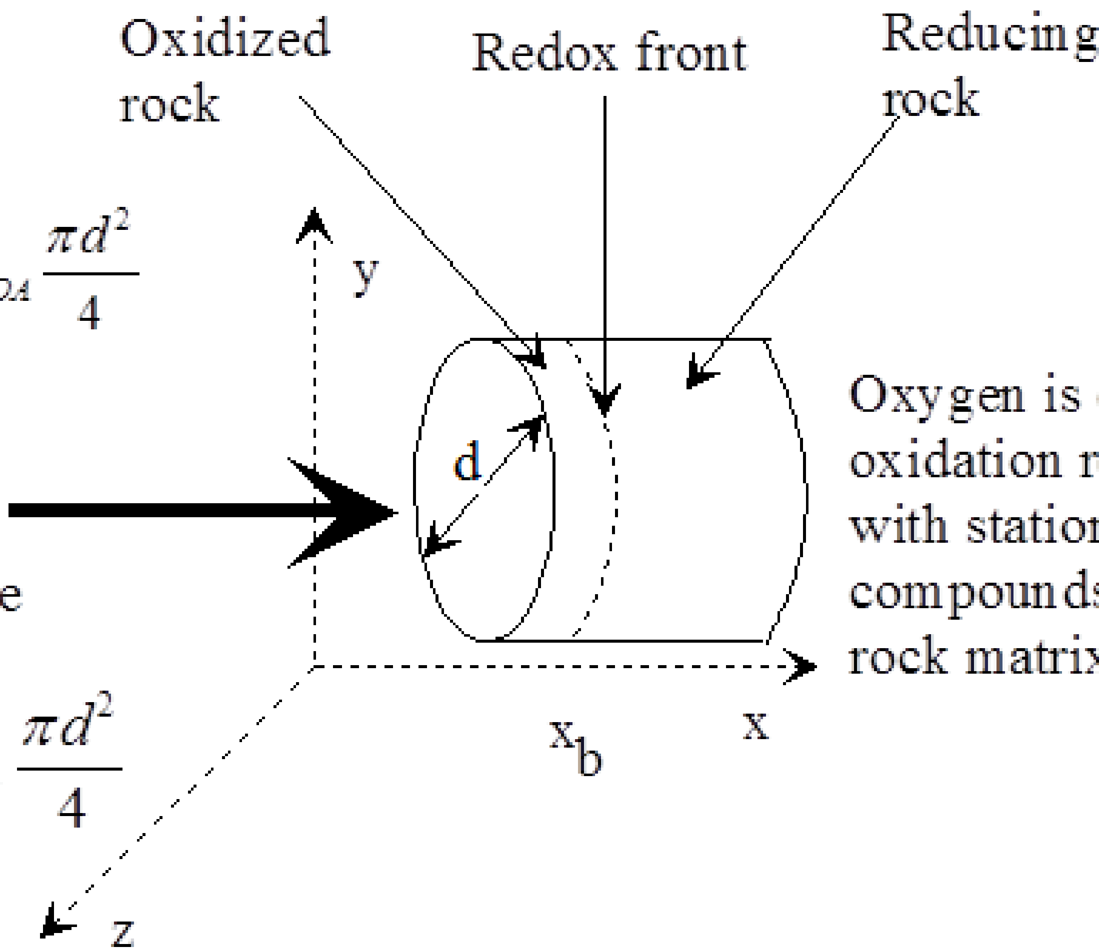
- Natural observations of concentration profiles in drill cores at Palmottu natural analogue site
- Drill cores were taken around water-carrying fractures
- Matrix diffusion was tested as the process to explain the observations
- Assumed driving force for U diffusion from rock matrix is the penetration of oxic water into bedrock that change conditions in fracture from anoxic to oxic
  - Glacial melt water pulses were tested
  - Diffusion of oxic water into rock matrix => U(IV) -> U(VI) => U mobilisation and subsequent release into fracture water
- U-series provide a tool for dating the release event(s)
  - U transport relative to immobile Th
  - Dating technique is ideal in the last glaciation time scale

Oxygen mass flow concept in drill core

$$\left.\frac{dN_{OA}}{dt}\right|_{adv} = -\frac{k \rho g}{\mu} \frac{\partial h}{\partial x} C_{OA} \frac{\pi d^2}{4}$$

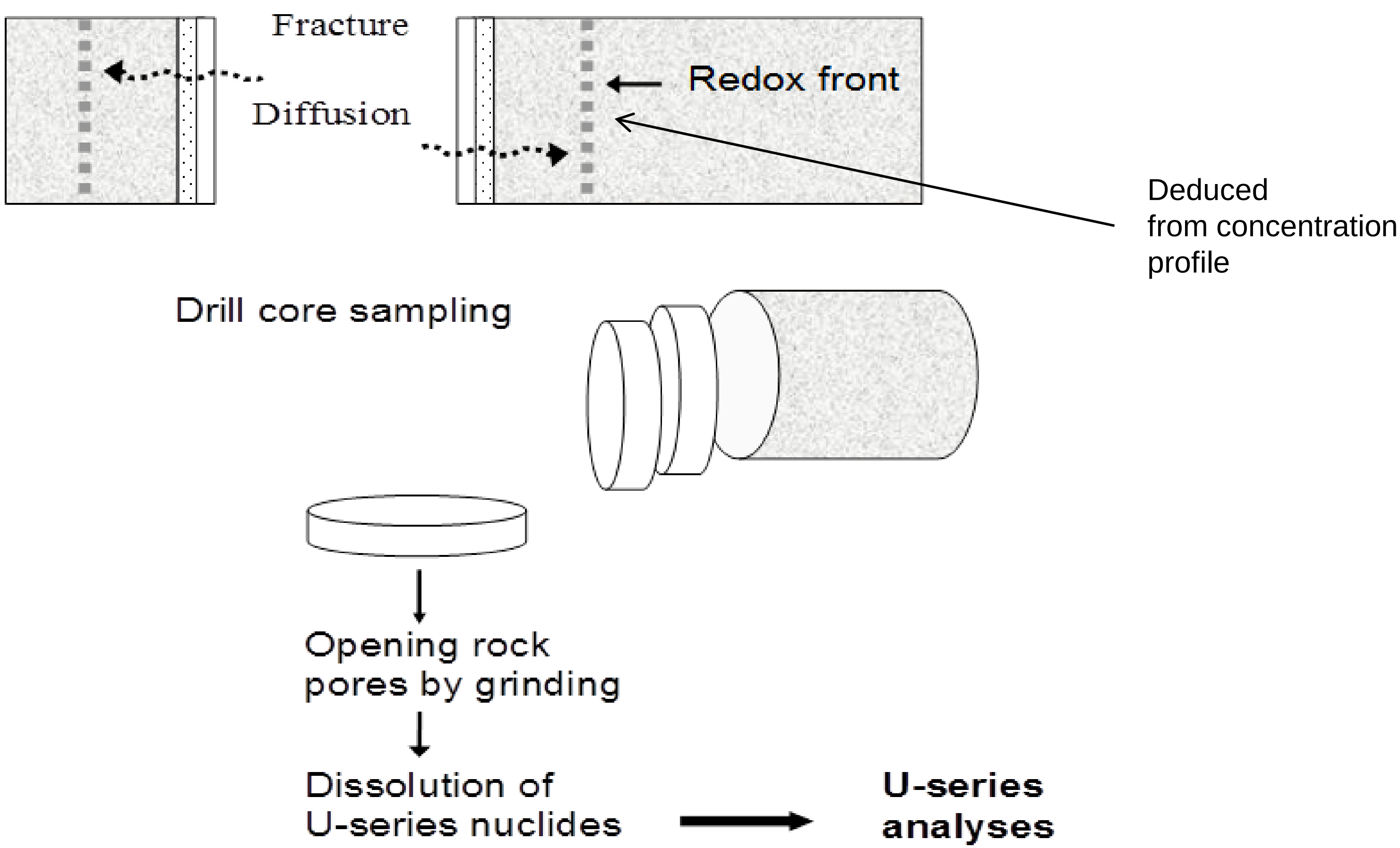
Mass flux of oxygen due to advection or diffusion from fracture

$$\left.\frac{dN_{OA}}{dt}\right|_{diff} = -\frac{dC_{OA}}{dx} D_e \frac{\pi d^2}{4}$$

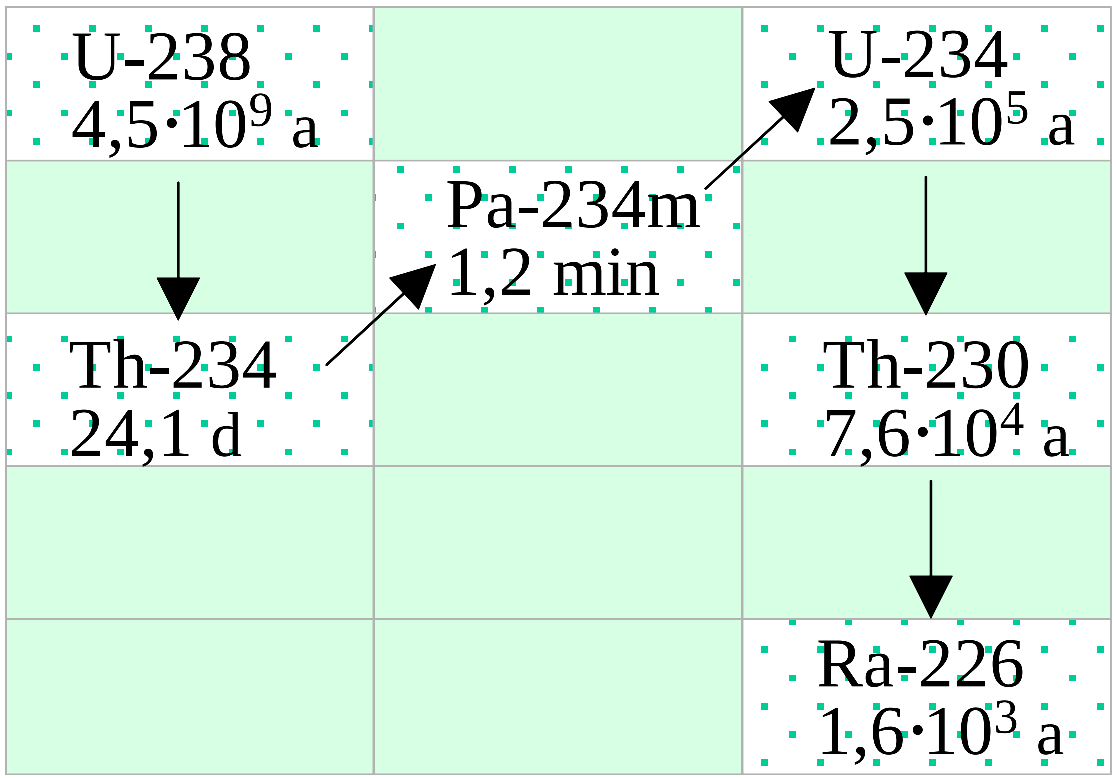


Oxygen is consumed in oxidation reactions with stationary Fe(II) compounds in the rock matrix

Sampling and analysis



Uranium-series (simplified)



Models used

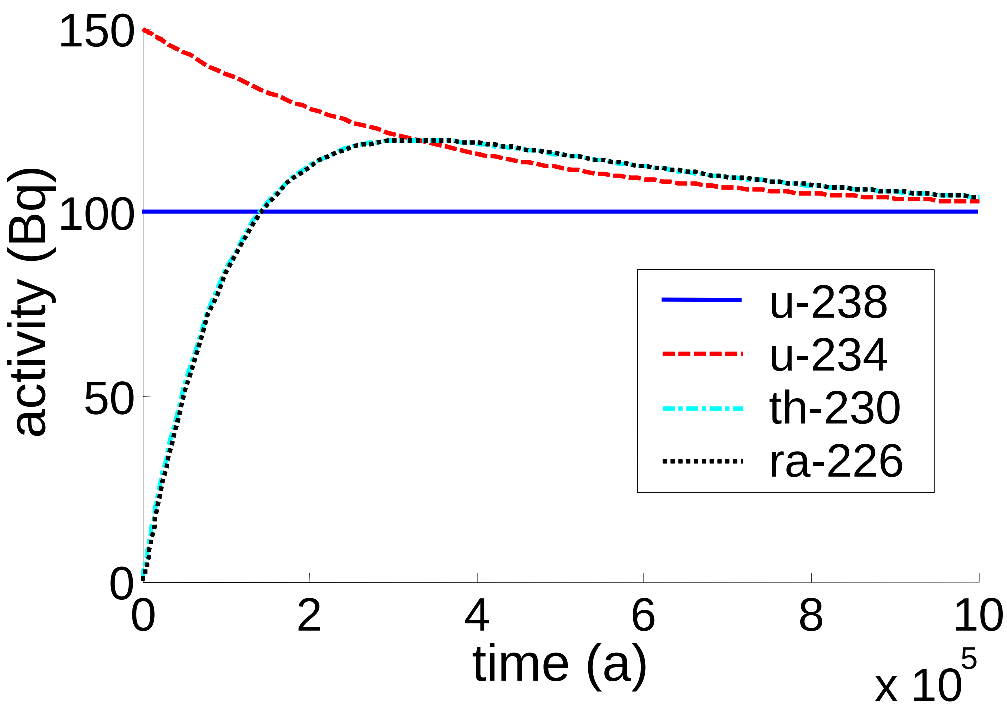
Uranium mass flow from rock matrix due to diffusion

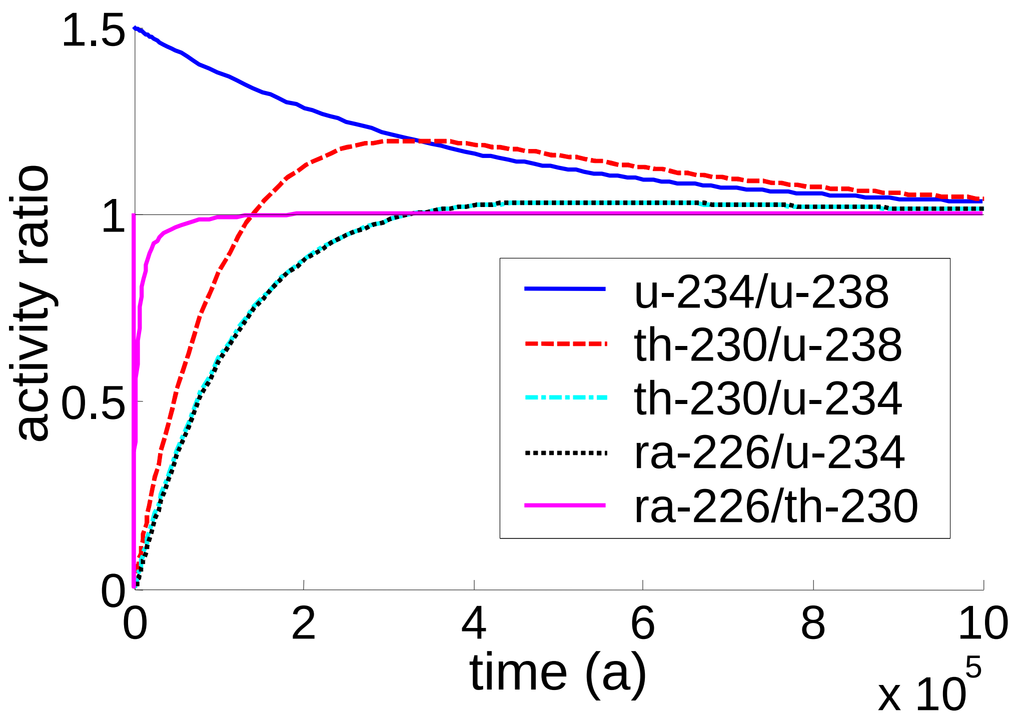
$$\frac{dN_U}{dt} = -D_e \frac{\pi d^2}{4} \frac{dC_U}{dx} \approx -D_e \frac{\pi d^2}{4} \frac{\Delta C_U}{\Delta x}$$

Uranium series disequilibrium model

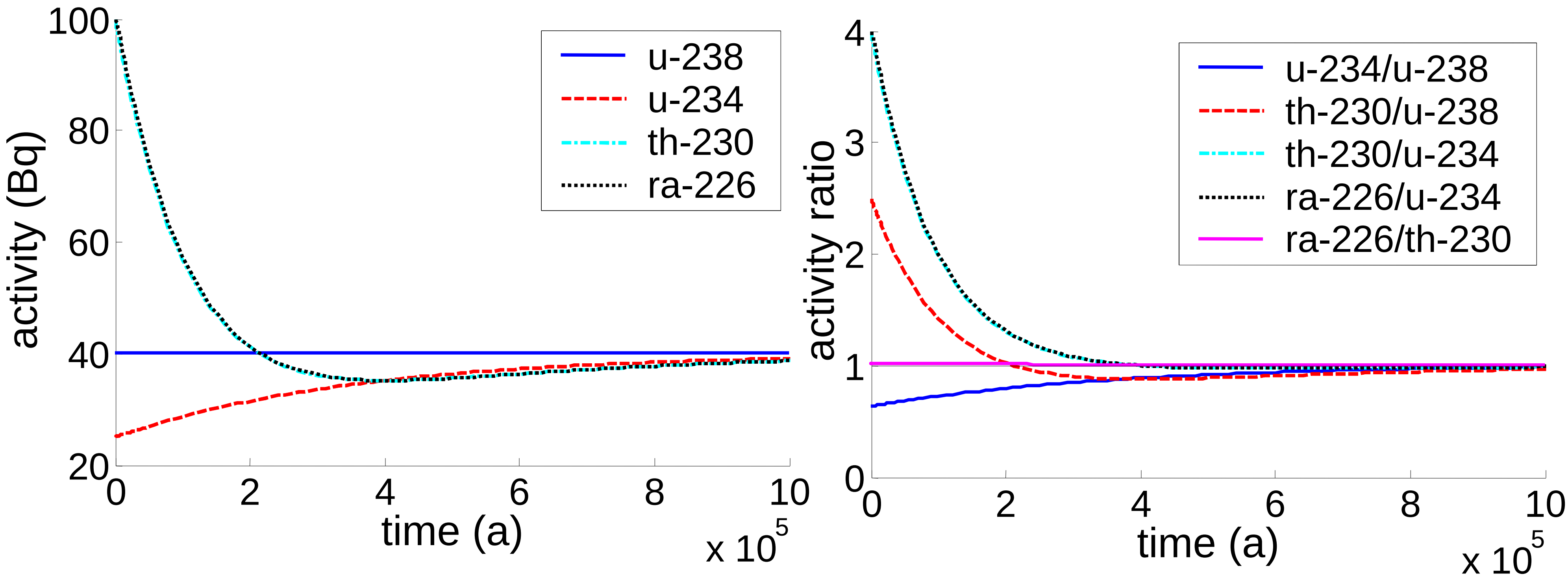
$$\frac{dC_i}{dt} = -\lambda_i C_i + \lambda_i C_{i-1} - R_i^- \ , \ i = 1, \ldots, 4, \ \ C_i(t_0) = C_i^0$$

Equilibration of U-series: sudden U accumulation
t=0: c0=0 Bq; t=1: U-238=100 Bq, U-234=150 Bq



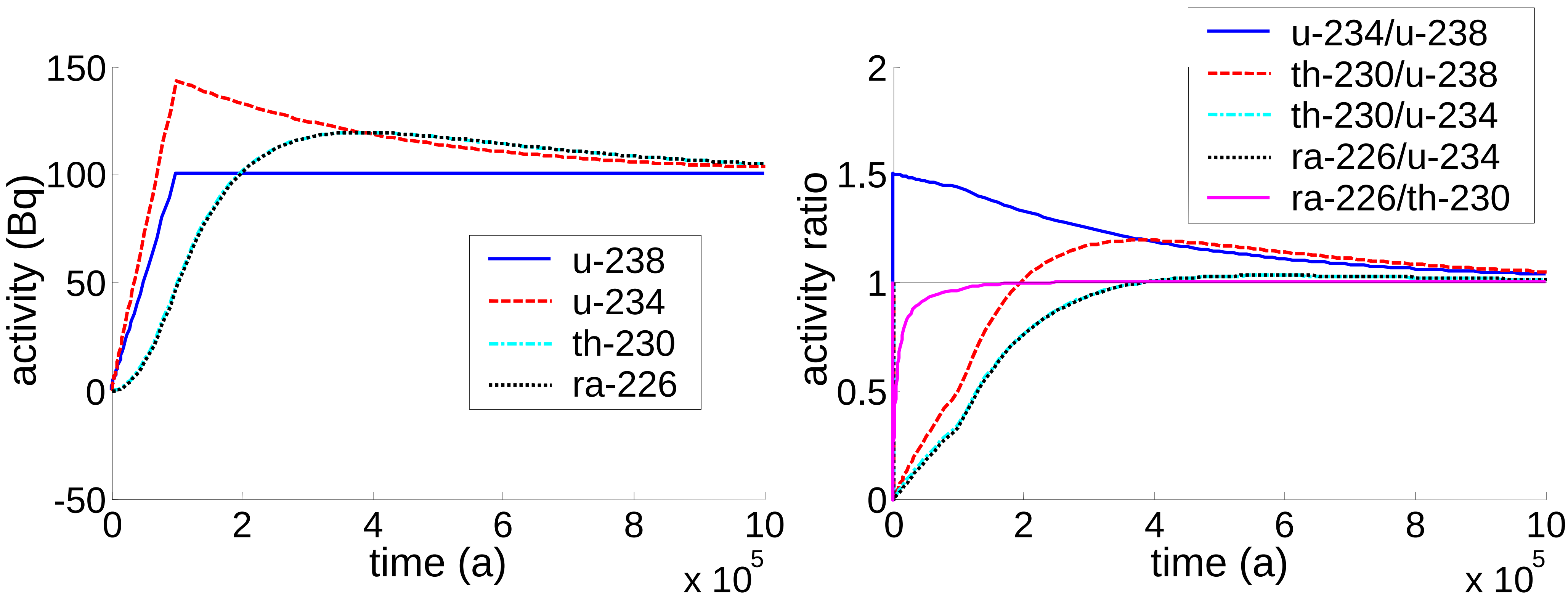


Equilibration of U-series: sudden U release
t=0: c0=100 Bq; t=1: U-238= 40 Bq U-234 = 25 Bq

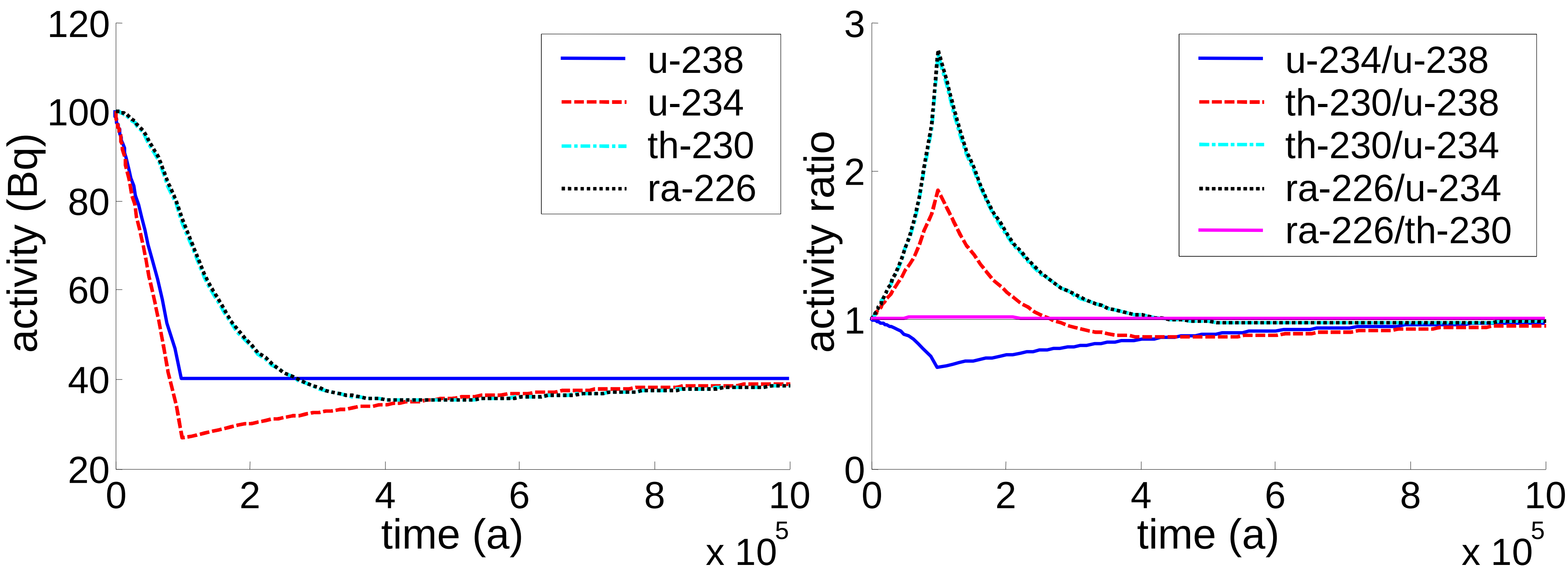


Equilibration of U-series: continuous U accumulation (constant accumulation rate for 1e5 a)

t=0: c0= 0 Bq; t=1: U-238-=-1e-3 Bq/a, U-234-=-1,5e-3 Bq/a



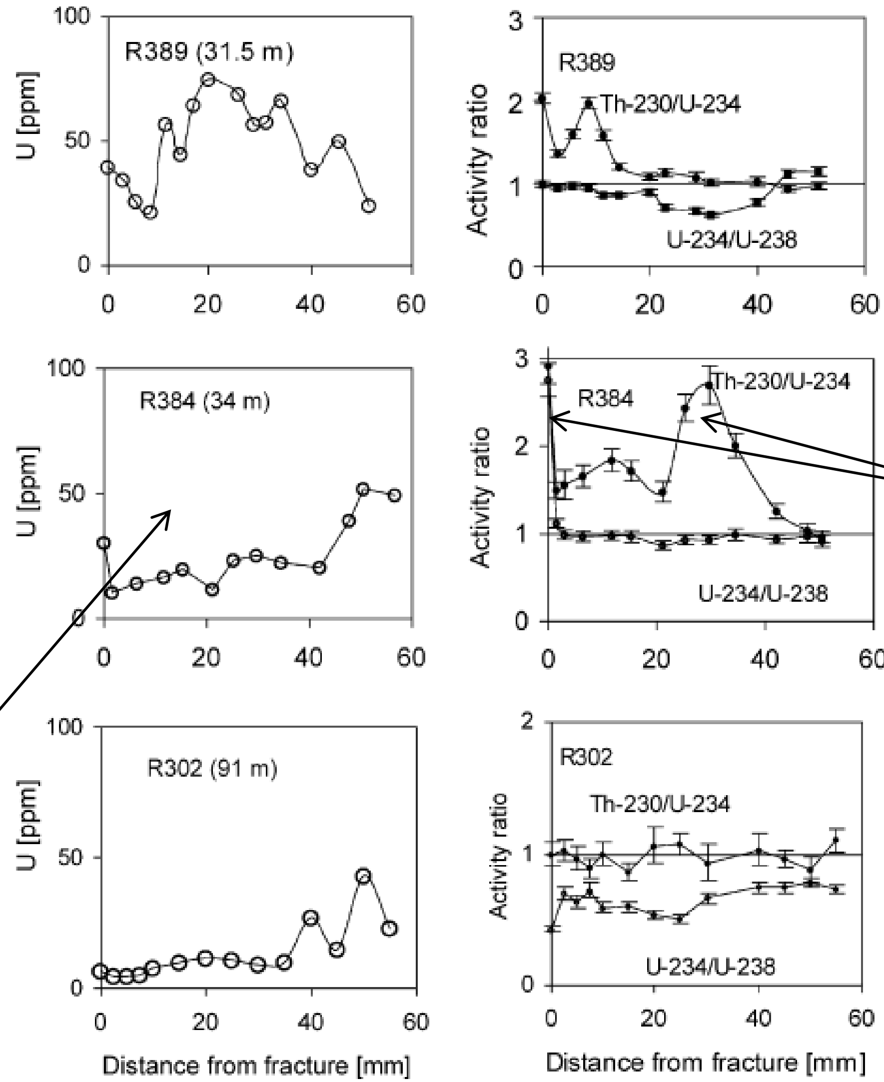
Equilibration of U-series: continuous U release (constant release rate for 1e5 a)
t=0: c0=100 Bq; t=1: U-238-=6e-4 Bq/a U-234-=7,5e-4 Bq/a



Observations

Profile studied

Missing U inventory



Recent release of U relative to immobile Th

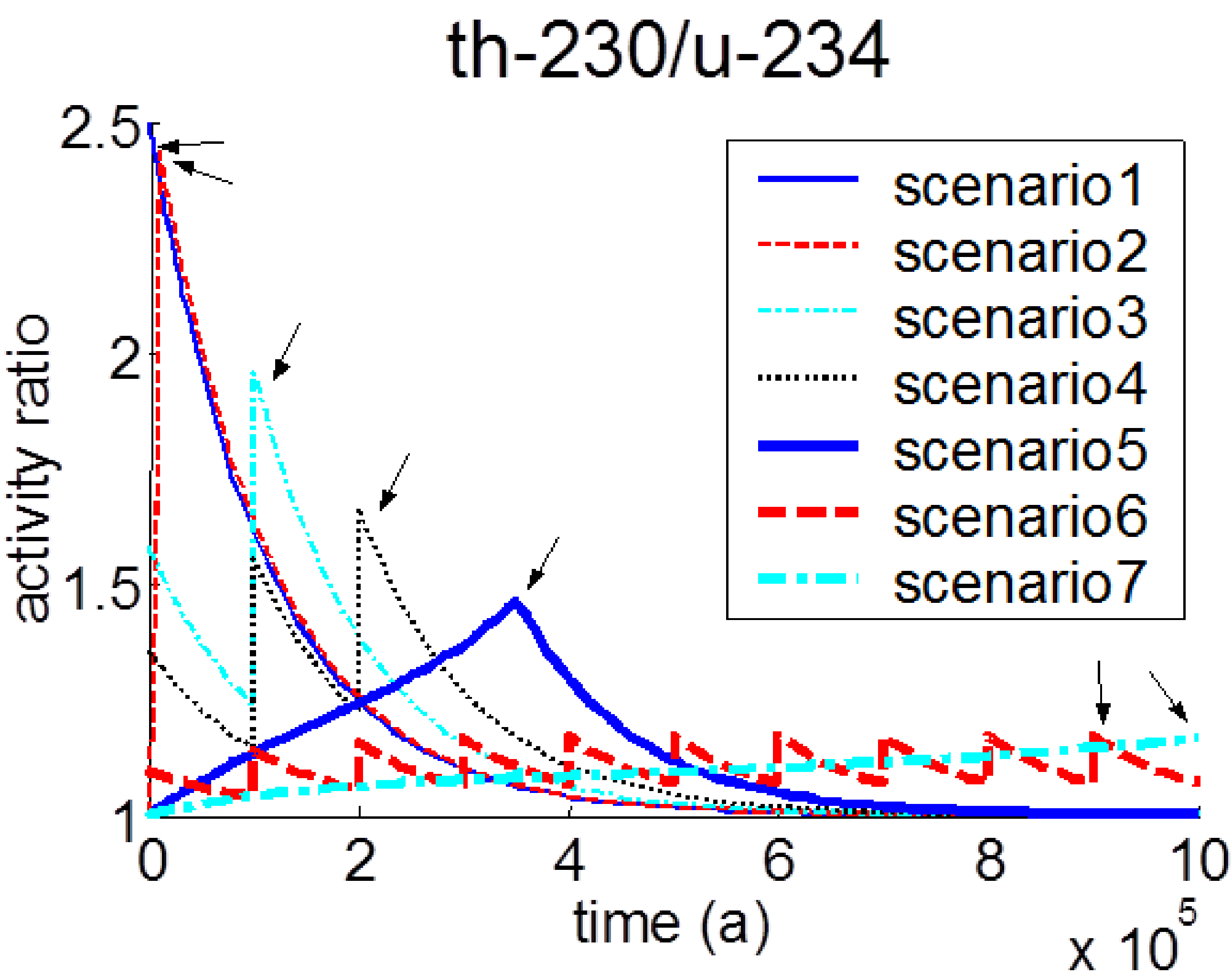
Assumptions tested

- Can standard matrix diffusion explain the ”missing U inventory” in the concentration profile
- How rapid has the U release been?
  - How fast can matrix diffusion transport the inventory?
  - What do measured uranium-series disequilibria tell about the time scales involved?
    - Release in one go?
    - Release in successive episodes?
      - Test latest glacial cycles as triggering events?

U release scenarios tested in USD simulations

Release scenario	Description	Conceptual assumption
1	Instantaneous release in one go	Instantaneous release at last deglaciation (10 ka ago)
2	Continuous release for 10 ka	Continuous constant rate release since last deglaciation
3	2 instantaneous release episodes 100 ka apart	Episodic release during two last deglaciations. Glacial cycle is assumed as 100 ka
4	3 instantaneous release episodes 100 ka apart	Episodic release during three last deglaciations
5	Continuous release for 350 ka	Continuous constant rate release for 350 ka (we know release has occurred during the last 350 ka)
6	10 instantaneous release episodes 100 ka apart	Episodic release during 10 consecutive deglaciations (reference to scenario 3)
7	Continuous release during 1 Ma	Continuous constant rate release for 1 Ma (reference to scenario 6)

Simulation results



Matrix diffusion constraint

- Calculated mass flow rate of U (the ”missing inventory”) from the drill core to fracture by matrix diffusion would take around 30 ky
  - => the U release cannot have occurred only after the last glaciation (around 10 k)

Final considerations

- Standard matrix diffusion and U-series disequilibrium modelling in parallel has given a plausible explanation for uraniun concentrations observed in a drill core
- Uranium series modelling provides possible release episodes that can explain the measured radioactive disequilibria
  - 4-5 possible episodes
- Matrix diffusion gives the minimum time (30 ky) the mass flow has to have been in action
  - This excludes single-event release after last glaciation
  - 2-3 possible release episodes left.
- Question not studied in detail: spreading of oxic conditions deeper into rock matrix
  - Redox front/boundary in the matrix
    - Importance in safety assessment?

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