Use of natural analogues to test the consistency of K_d values in performance assessment

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Background

- In SKB's safety assessment SR-Site (2010), a simplified model of steady-state radionuclide transport was used to check consistency of predictions of ^{226}Ra & ^{222}Rn activity with groundwater measurement data.
- In the work presented here, a more detailed decay-chain transport model, **MC23** (Mahmoudzadeh & Crawford, 2023) was used to simulate natural radionuclide transport in fractured rock.

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Modelling simultaneous transport of natural and anthropogenic radionuclides in fractured media — Diffusion into the heterogeneous layered rock matrix for an arbitrary length decay chain

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Are Smart K_d estimates for ^{226}Ra consistent with long-term field observations?

Smart K_d concept…

- Based on the premise that relative variations in modelled K_d values should mirror the variation in "true" (measured) values:

"best estimate" K_d for reference groundwater

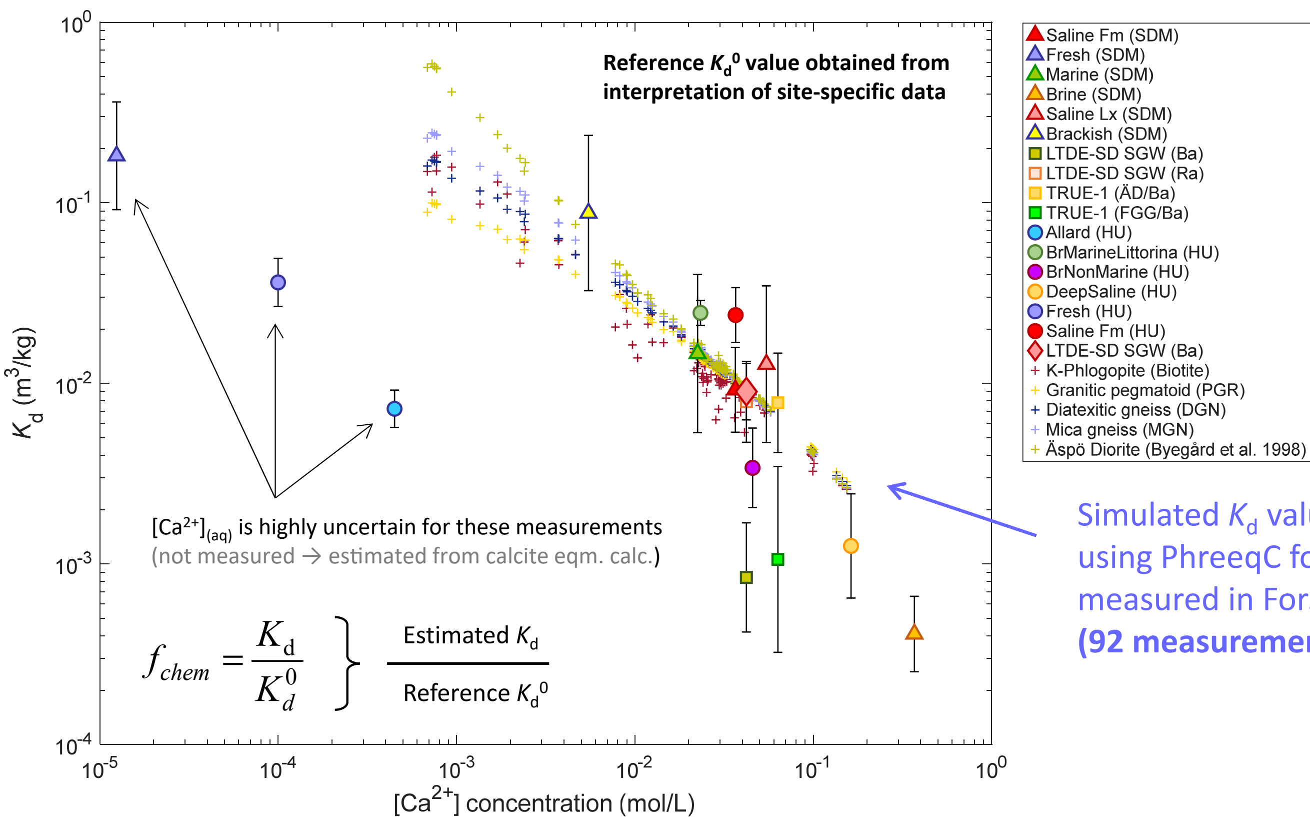
$$\frac{K_{\mathrm{d}}}{K^{0}_{\mathrm{d(meas)}}} = \frac{K_{\mathrm{d}(mdl)}}{K^{0}_{\mathrm{d}(mdl)}} = f_{chem} \Rightarrow K_{\mathrm{d}} = f_{chem} \cdot K^{0}_{\mathrm{d(meas)}}$$

chemistry transfer factor for application groundwater

- f_{chem} is calculated using a mechanistic model (many choices available, SC/CE).
- Underlying assumption is that uncertainty in model parameters is reduced by taking the ratio of modelled K_d values (i.e. mutual cancellation of uncertainties).
- The challenging part is to estimate a reliable reference K_d^0 from measurement data.

Proposed Smart K_d model

- In multiple candidate models tested (both cation exchange & surface complexation), a clear dependency of K_d on the total Ca^{2+} concentration is observed (adsorptive competition between Ra^{2+} and Ca^{2+})



Simulated K_d values ("+" symbols) calculated using PhreeqC for groundwater compositions measured in Forsmark boreholes

(92 measurements in different BH sections)

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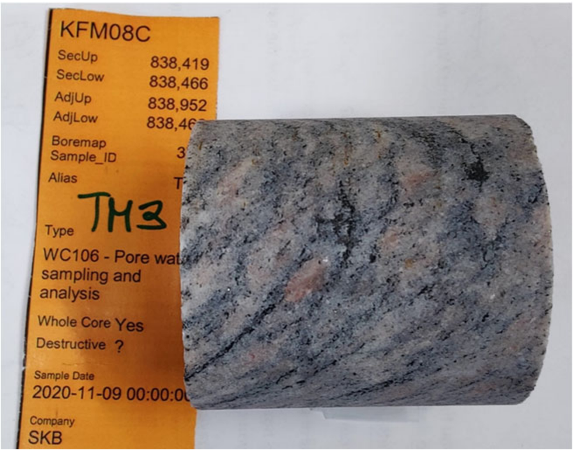
Transport model parameters

Site-specific inputs:

- K_d for Ra sorption (fracture minerals & rock matrix)
- matrix emanation factor (recoil capture fraction)
- fracture transmissivity (transport aperture)
- rock bulk density & porosity
- matrix formation factor (effective diffusivity)
- U & Th content of rock & fracture minerals
- fracture mineral abundance (g/m^2)
- Ba-content of fracture minerals (chlorite & barite)
- background $[^{238}U]_{(aq)}$ concentration

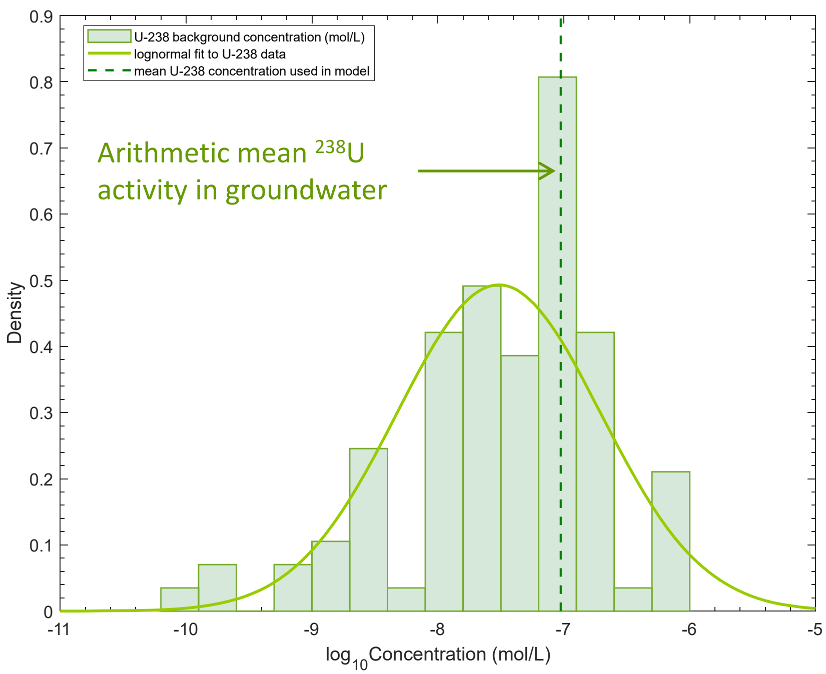


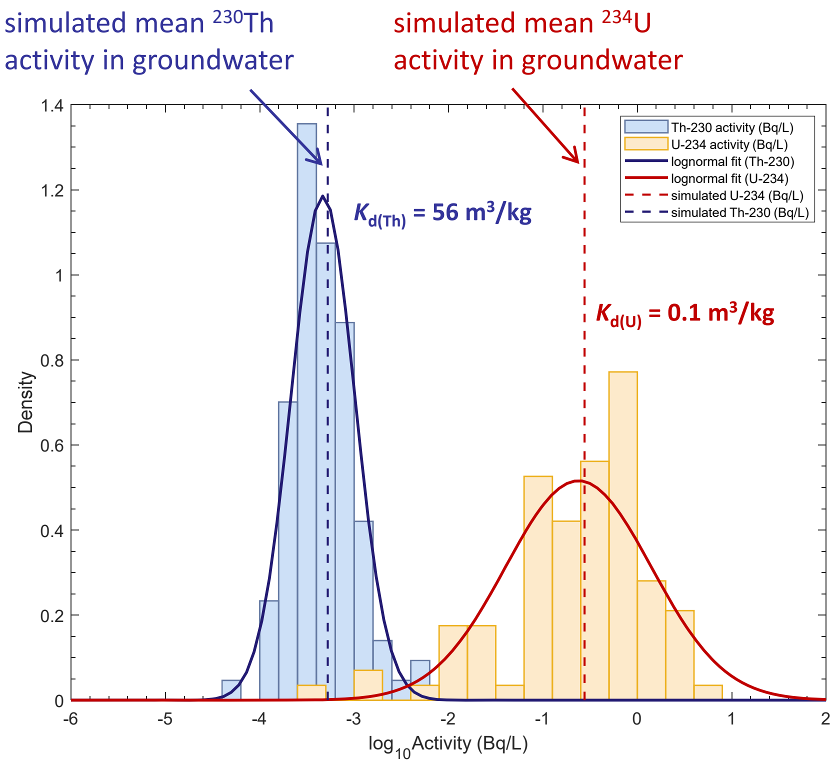
Assumed representative values:

- K_d for U & Th sorption (calibrated to aqueous concentrations)
- fracture mineral emanation factor (recoil capture fraction, 50%)
- Ba-mass fraction as $BaSO_4$ in fracture minerals (limiting case, 100%)
- matrix diffusion depth (effectively infinite on timescale of isotopic equilibrium)
- advective travel time ("old" groundwater, t_w > 50y)
- fracture mineral sorption enhancement factor relative to fresh rock (≈ BET surface area ratio)

Transport model parameters (cont'd)

- ^{238}U background is specified as an input parameter directly from the measurement data
- K_d for ^{234}U and ^{230}Th adjusted to give rough (visual) fit to measurement data





Calibration was limited to U/Th background behaviour; Ra predictions remain independent.

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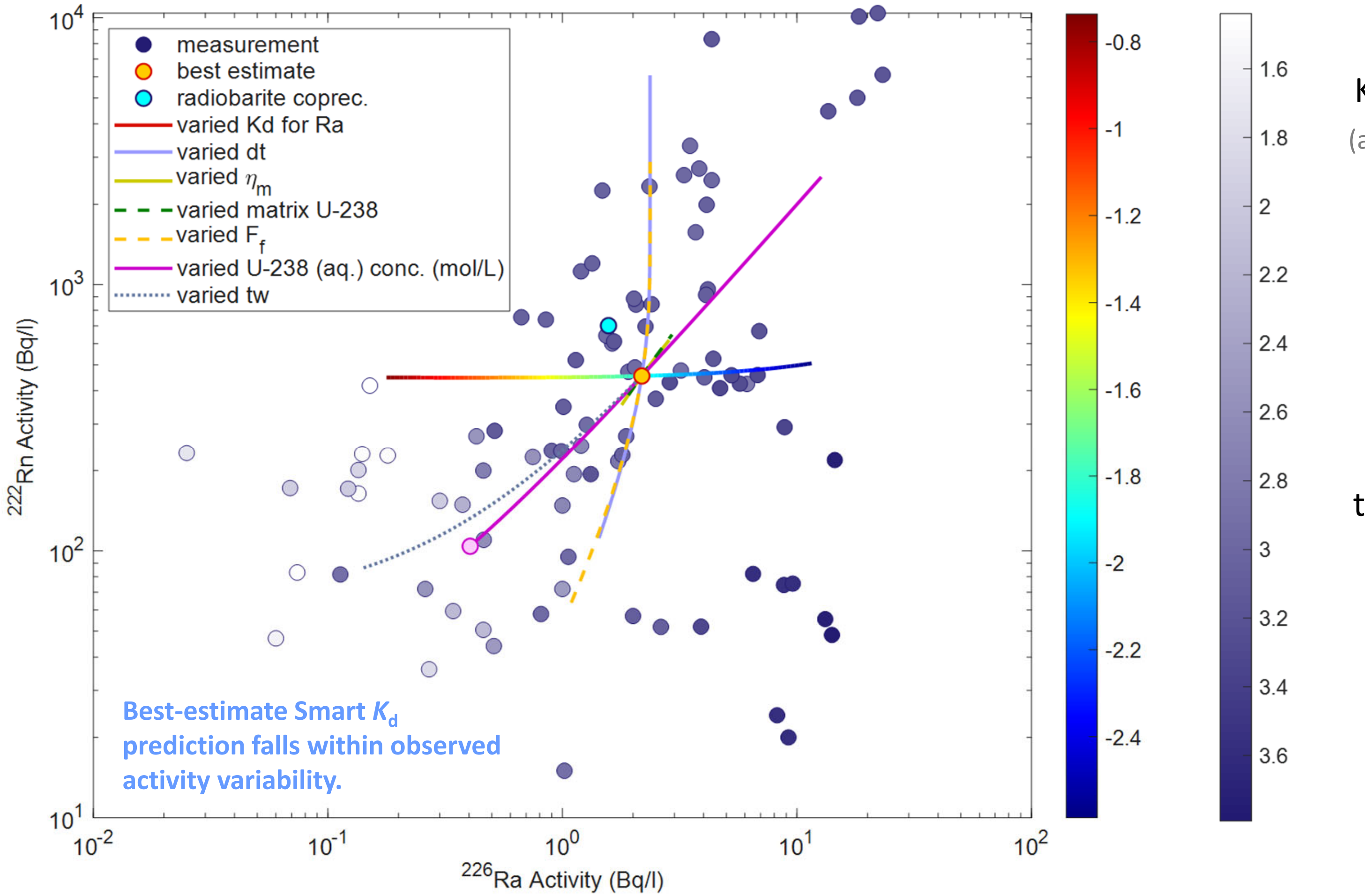
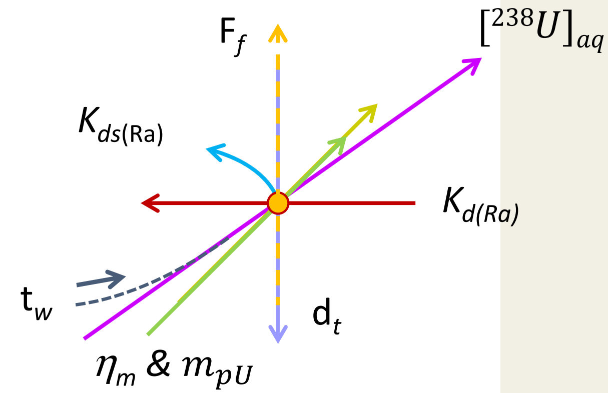
Results for Forsmark measurements (mean properties)

Data available for 92 borehole sections at Forsmark



Key to variation analysis…

(arrows indicate direction of increase)



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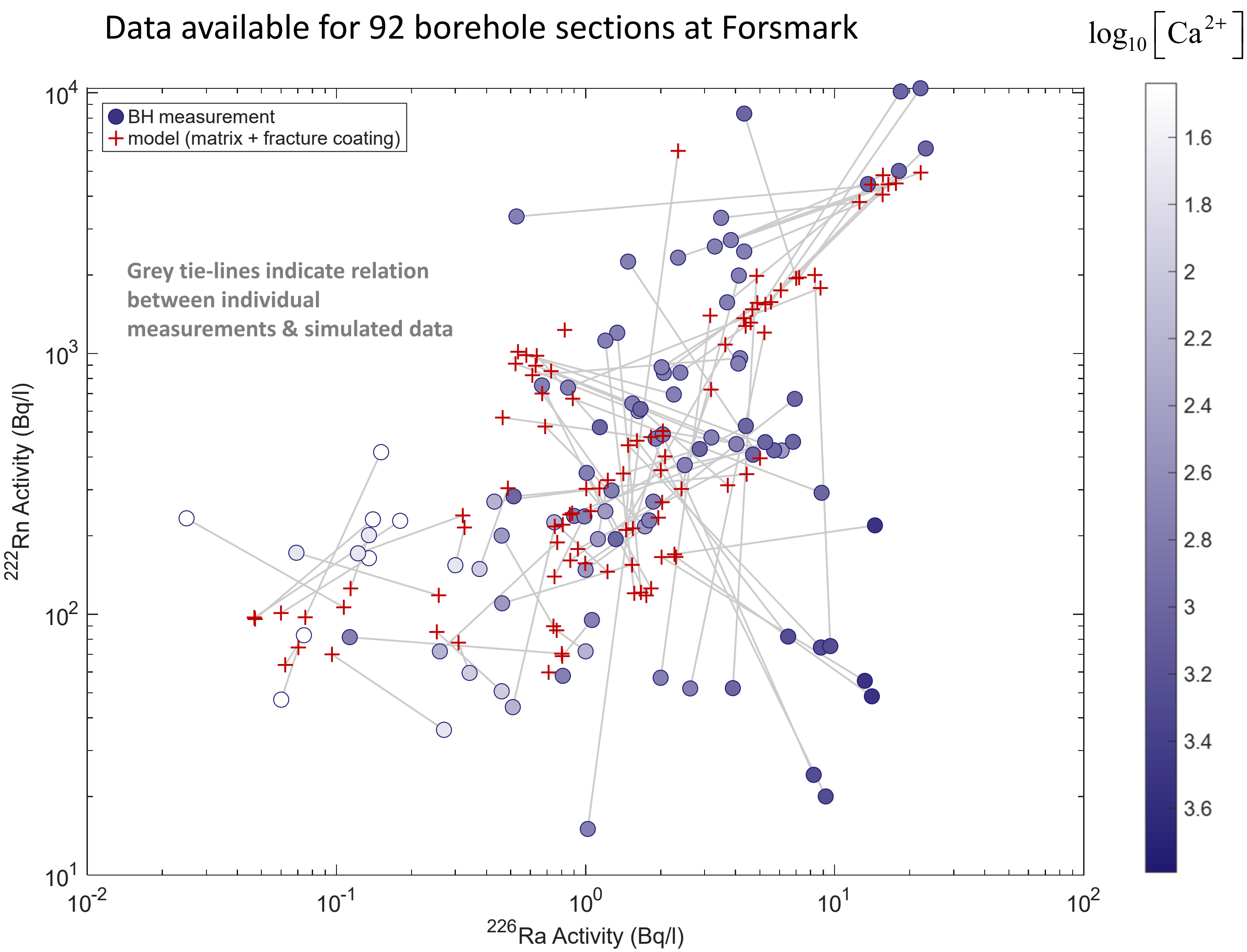
Results for Forsmark measurements (BH-section specific)

Data available for 92 borehole sections at Forsmark



BH-section specific variables:

- **transport aperture**
- **rock matrix $K_{dm(Ra)}$**
- **fracture coating $K_{ds(Ra)}$**
- **background $[^{238}U]_{(aq)}$**

All other variables assigned representative “average” values

Predictive uncertainty is approximately half an order of magnitude for both ^{226}Ra and ^{222}Rn (~ ±0.4 log units)

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Conclusions

- Independent K_d values for Ra, adjusted using a chemistry-dependent transfer factor, are broadly consistent with observed ^{226}Ra and ^{222}Rn activities in the Forsmark natural analogue.
- Borehole section-specific agreement is encouraging (~0.4 log units on average), although substantial unexplained variability remains.
- The results provide encouraging support for the Smart K_d concept, although additional testing against independent datasets would be valuable.
- The results suggest that transport and hydrogeological uncertainties may now be more important than uncertainties associated with the Smart K_d transfer factor approach.
- Porewater capture of radionuclides is very low for unaltered Forsmark rock (~3.5%) and requires significant contribution from decay of mobile ^{238}U to explain ^{226}Ra & ^{222}Rn activity levels.
- The calculations suggest that it may be also possible to check consistency of independently estimated K_d values for ^{234}U and ^{230}Th using the MC23 transport model (**future goal = no calibrations**).

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(Extra material) Effective penetration depths…

Average depth in rock matrix to/from which a radionuclide will diffuse before undergoing decay:

pore diffusivity (m^2/s)

$$\delta_p = \sqrt{\frac{D_p}{\lambda R_{pm}}}$$

decay constant (1/s)

pore retardation factor (-)

Crawford & Löfgren, 2019 (SKB R-17-22)

Neretnieks, 2013 (10.1007/s10040-013-0986-z)

$$\delta_{p(U-238)} \approx 1.55 \text{ m}$$

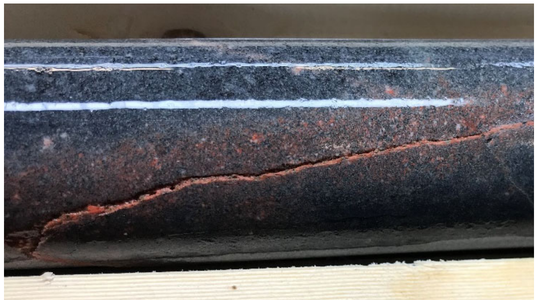
$$\delta_{p(U-234)} \approx 1.15 \text{ cm}$$

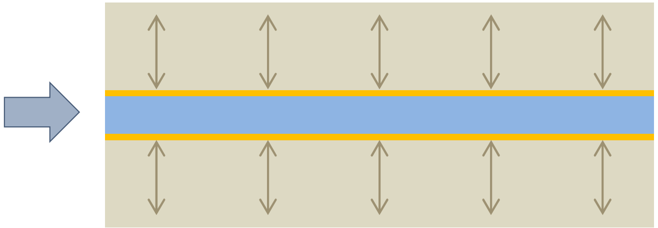
$$\left.\begin{aligned} &\delta_{p(\mathrm{Th}-230)} \approx 0.3 \text{ mm} \\ &\delta_{p(\mathrm{Ra}-226)} \approx 3 \pm 1 \text{ mm} \\ &\delta_{p(\mathrm{Rn}-222)} \approx 1 \text{ mm} \end{aligned}\right\}$$

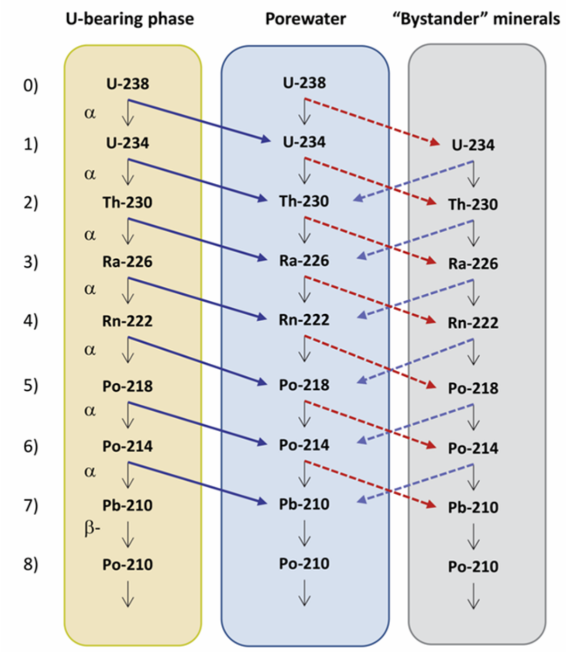
Could be as much as an order of magnitude larger if D_p is 100× greater in rim zone

The short penetration depths predicted for radionuclides in the U-series imply that natural analogue observations are unlikely to provide a strong test of matrix diffusion.

(Extra material) MC23 natural analogue model







Processes considered:

- Natural production of radionuclides (recoil capture from mineral grains).
- α-recoil loss of radionuclides from porewater to "bystander" minerals (usually neglected in PA).
- Advective transport & matrix diffusion.
- Sorption on mineral surfaces in rock matrix.
- Enhanced sorption on fracture coatings (including radiobarite).
- Full decay chain mass balance including background ^{238}U (aq.) in porewater.
- Possibility to include an arbitrary number of alteration layers adjacent to the fracture surface (neglected in present calculations).

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