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Effects of the hydrogeochemical variability of pore water in the Opalinus Clay and its surrounding aquifers on uranium migration

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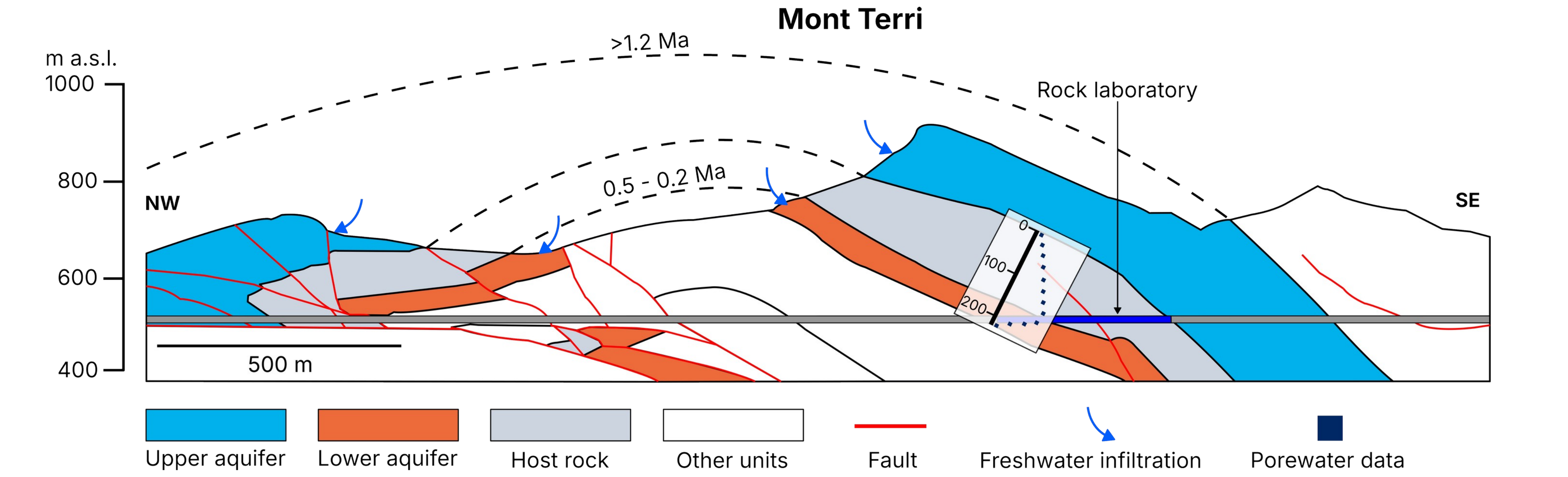
Motivation

How does the hydrogeochemical variability of boundary and initial conditions affect uranium migration through host rock?

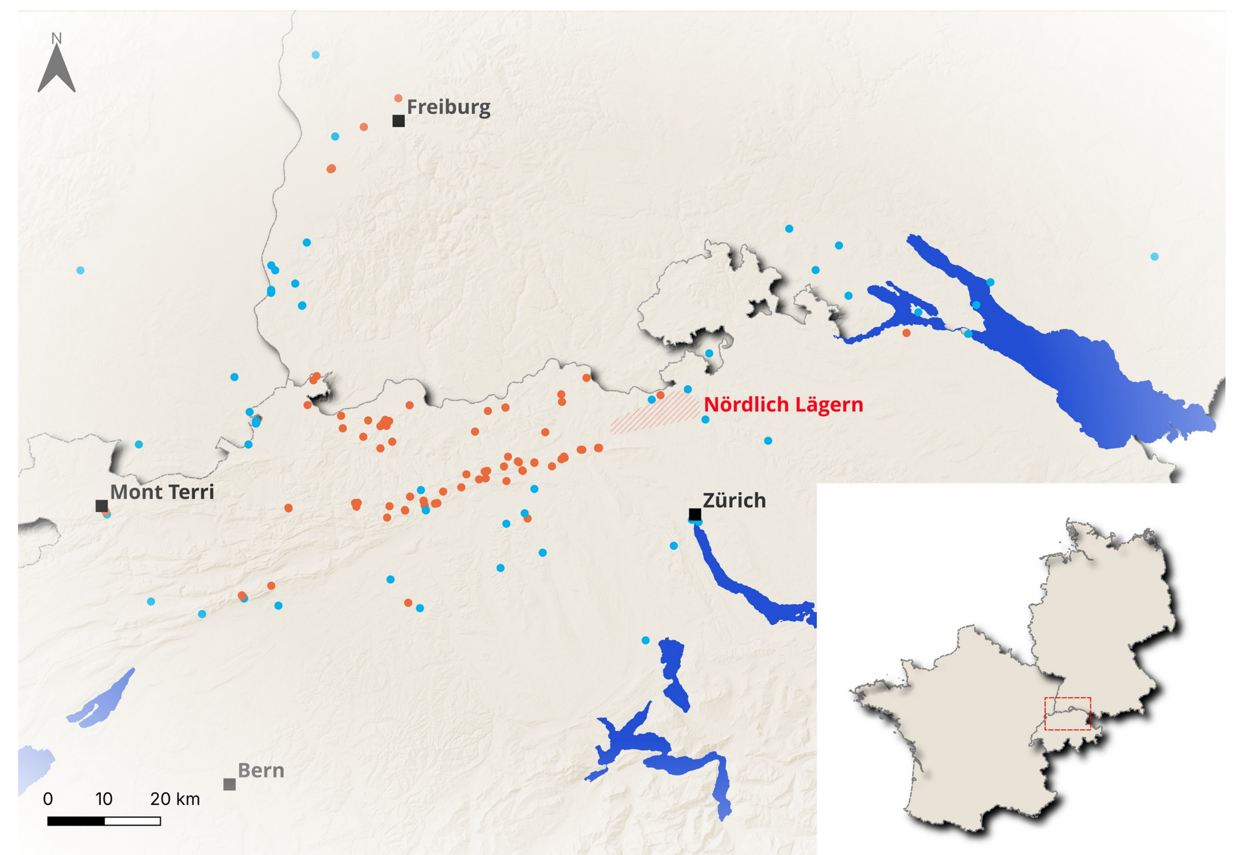
- Pore water and groundwater data are required to define initial and boundary conditions for simulations of radionuclide migration for safety assessments
- Hydrogeochemical compositions of host rock and adjacent aquifers are subject to spatial and temporal changes with potential effects on uranium migration [1]
- Opalinus Clay is chosen host rock for Swiss nuclear waste disposal, has been well studied at Mont Terri and is also relevant for German site selection process



Erosion history of Mont Terri anticline and formation of the present hydrogeological system. Freshwater infiltration into the upper and lower aquifers surrounding the host rock formation Opalinus Clay caused the development of a gradient in pore water hydrogeochemistry [2]. Modified from [3].

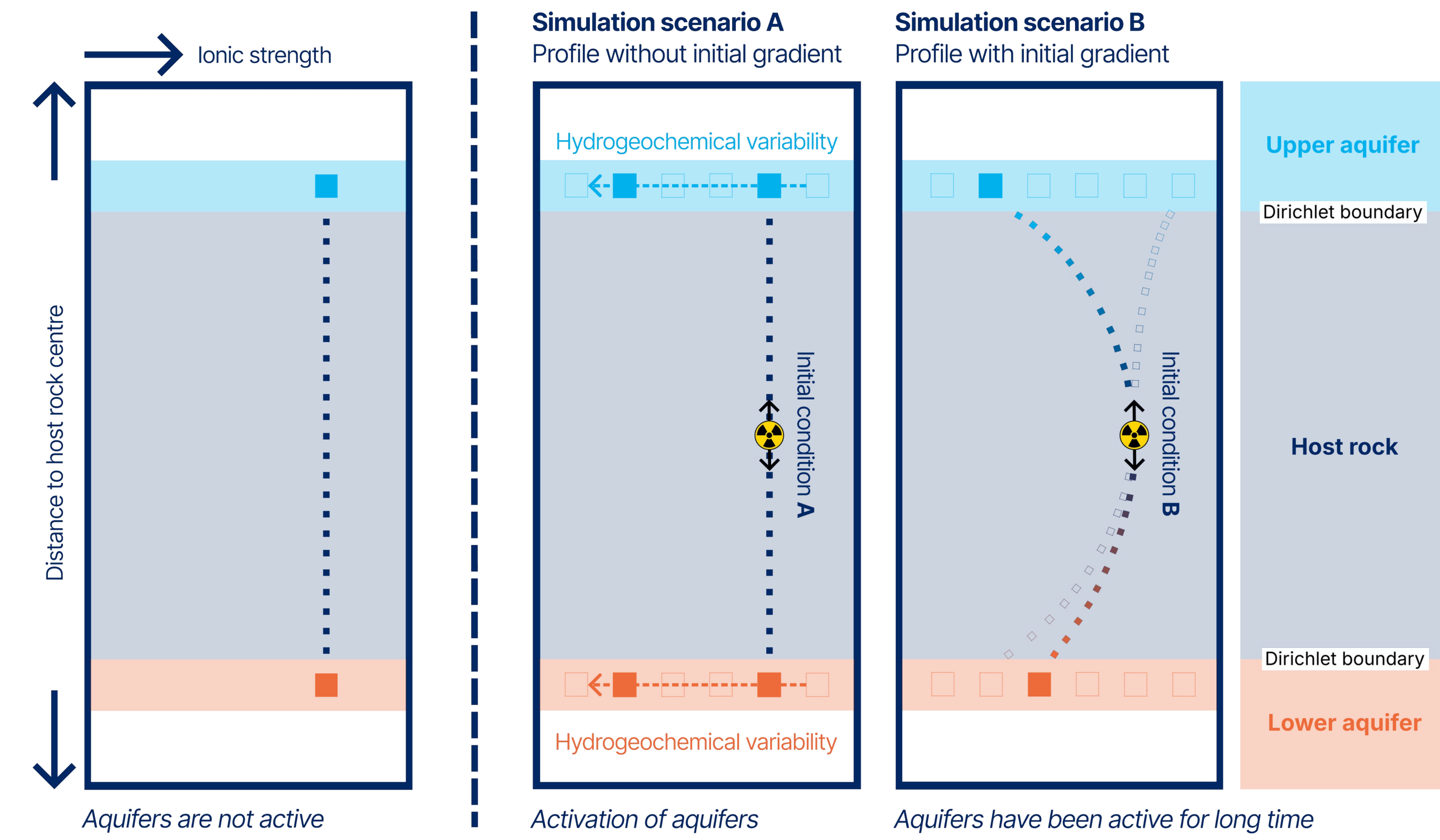
Methods

Reactive transport simulations with varying aquifer compositions at model boundaries and different initial profiles of pore water hydrogeochemistry



Locations of groundwater samples from border triangle of Switzerland, Germany and France (blue: upper aquifer; orange: lower aquifer). The red hashed area indicates the proposed Swiss nuclear waste disposal site.

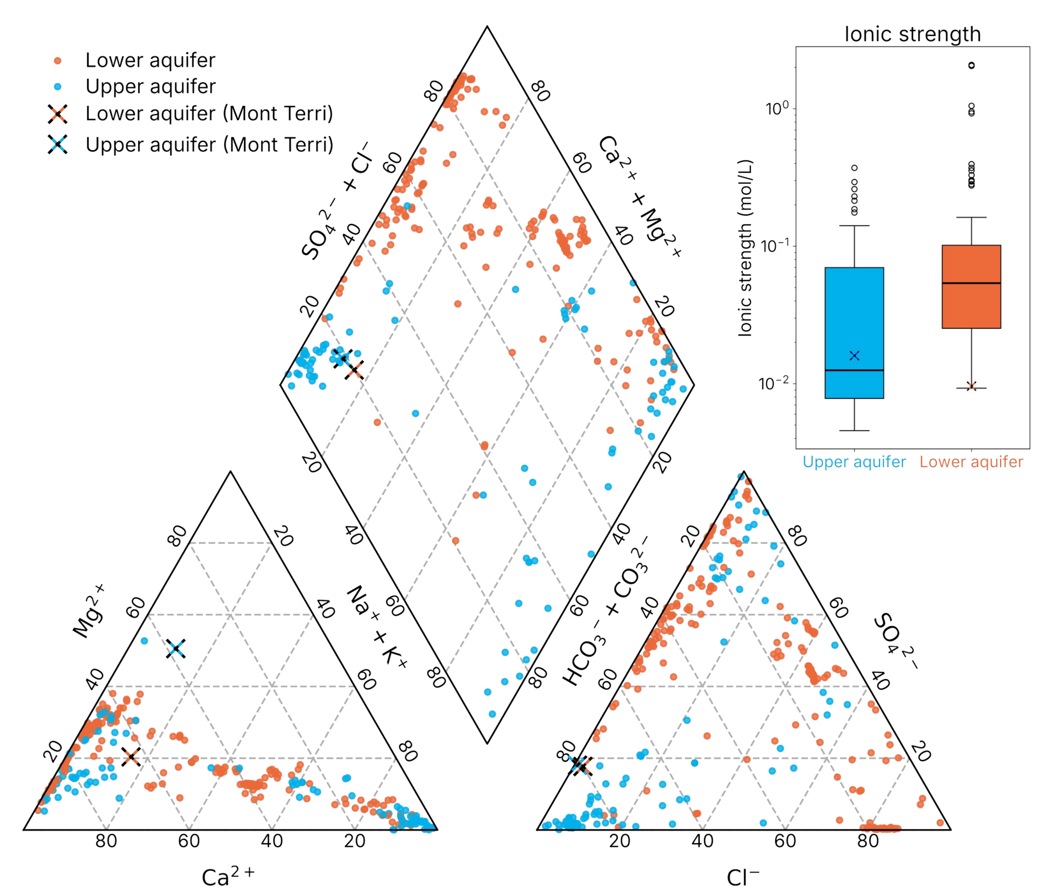
- 212 samples from aquifers in Molasse basin compiled [4, 5]
- Each groundwater composition applied as a boundary condition
- Present-day hydrogeochemistry at Mont Terri as reference case
- 1D model in PHREEQC [6] with simulations over one million years
- Fick's diffusion, cation exchange, surface complexation considered
- Mineral solution equilibria with pyrite, siderite, calcite, dolomite



Schematic illustration of study concept. Boundary conditions are varied in two scenarios (A, B) that differ in initial profiles of pore water hydrogeochemistry. The nuclear waste repository is represented by a constant uranium source term in the model centre. Modified from [7].

Results

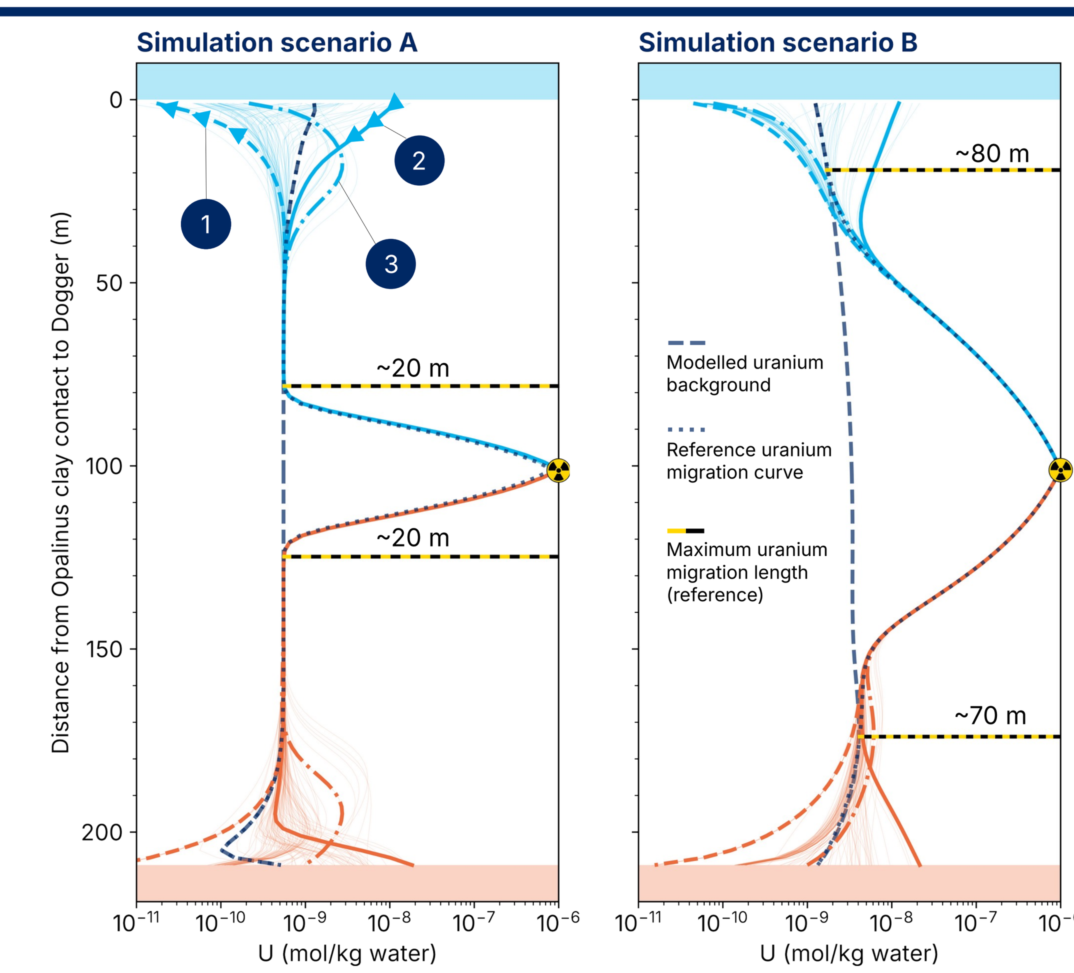
Uranium migration lengths through Opalinus Clay differ by several decametres depending on initial profile of pore water composition



Piper-plot indicates broad spectrum of water types in the investigated groundwater samples. Mean value and range of ionic strengths are higher in the lower aquifer than in the upper.

- Similar ionic strengths of groundwaters in northern Switzerland and southern Germany [8]
- Effects on uranium concentration at model boundaries can be summarised as follows:

1. lower natural uranium concentration in aquifers
   → *uranium diffuses out of the model*
2. Higher natural uranium concentration in aquifers
   → *uranium diffuses into the model*
3. higher alkalinity in aquifers
   → *dissolved uranium increases in the model*



Hydrogeochemical variations of surrounding aquifers influence natural uranium concentrations close to the model boundaries, but not migration from the repository in the centre over 1 Ma.

Conclusions

Hydrogeochemical variability of initial pore water composition is more decisive for uranium migration than variability of boundary conditions

- Hydrogeochemical gradients in pore water of host rock increase uranium migration lengths [9]
- Variable aquifer compositions only affect natural uranium background over one million years
- Pore water data required at local level, while regional data on aquifer compositions are sufficient
- Constant pore water hydrogeochemistry across host rock is to be favoured for disposal sites
- Findings are relevant for similar argillaceous formations and the German site selection process
- Future simulations need to consider multi-component diffusion and other radionuclides

References

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