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Supplement of

Impact of the heterogeneity of the sandy Opalinus clay facies at the Mont Terri underground research laboratory on radionuclide migration

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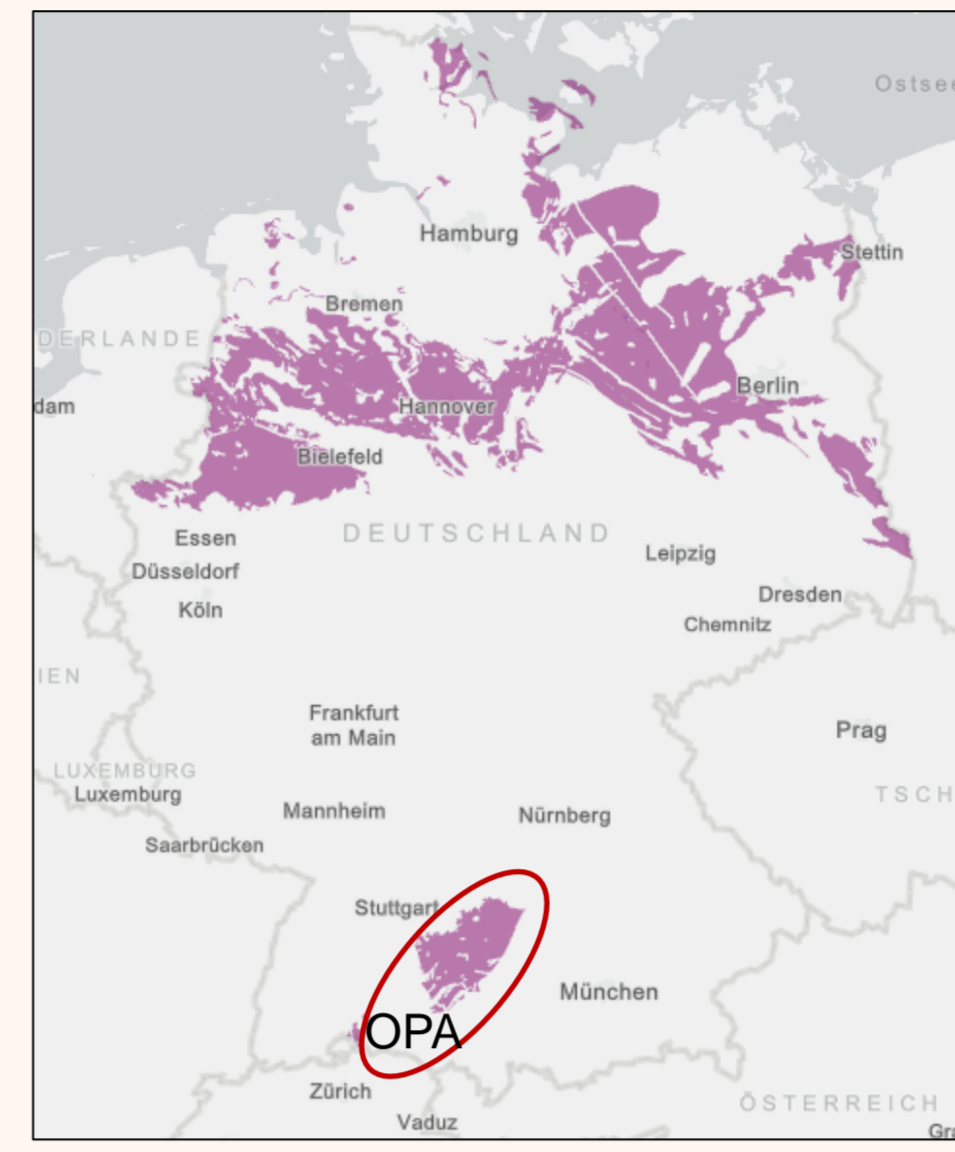
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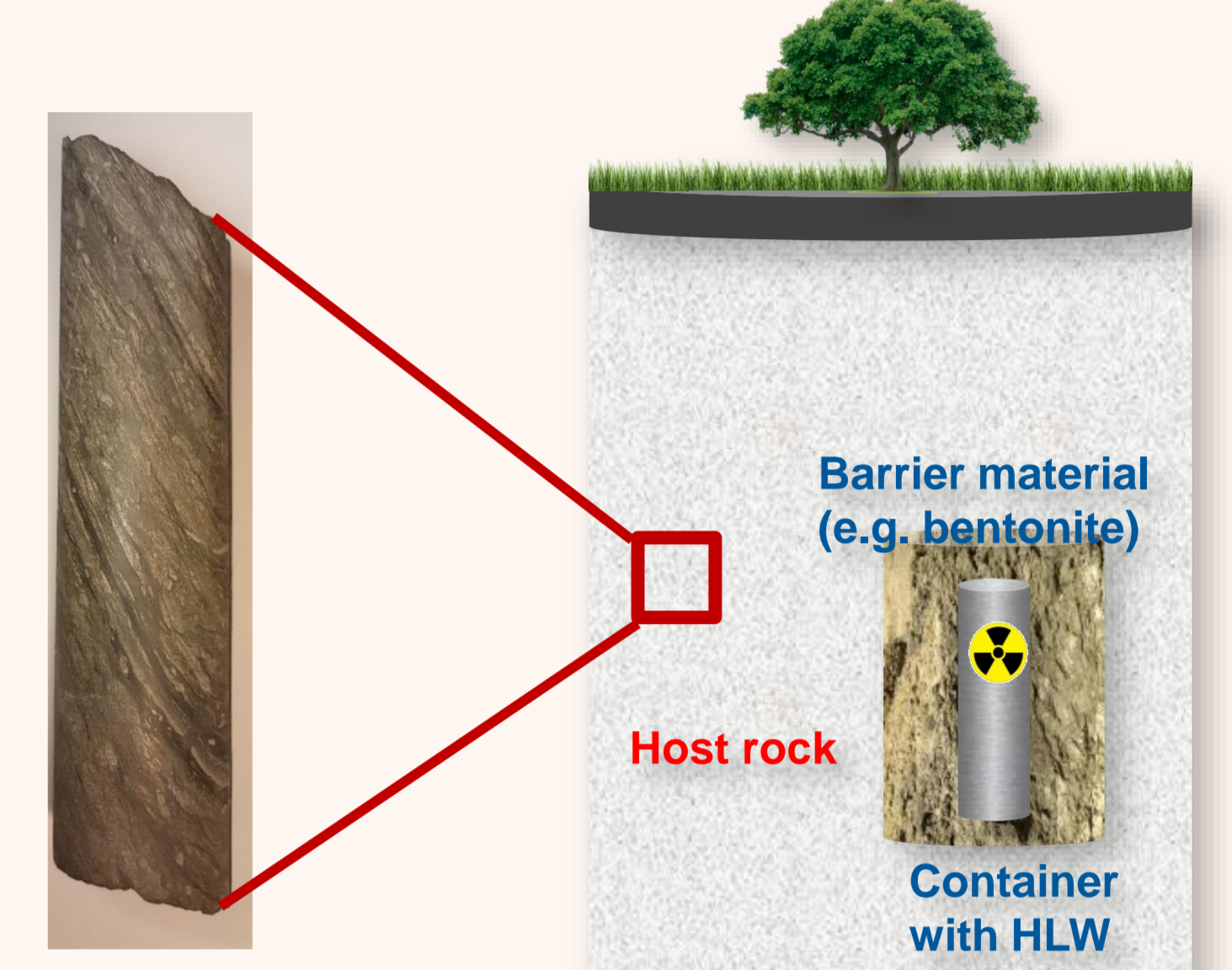
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Background / Objectives

- Within the site selection procedure for a repository for heat-generating, high-level radioactive wastes, the site with the "best-possible safety" is to be determined in a multi-phased comparative process.
- Potential host rocks for nuclear waste repositories are heterogeneous on various scales, ranging from nano-scale to kilometer-scale.
- Objective: Evaluation of the effects of structural and mineralogical heterogeneity of a clay host rock on ²²⁶Ra (half-life 1600 years) migration using sandy Opalinus Clay (OPA) facies as example.



Pretertiary clay rocks identified in the "Zwischenbericht Teilgebiete" (www.bge.de)



Drill core extracted from Mont Terri underground laboratory

Tools / Methods

Chemical / structural characterization:

e.g., XRD, SEM-EDX, N₂-BET, quantitative Rietveld phase analysis using GSAS-II software.

Determination of transport parameters:

Determination of HTO effective diffusion coefficient D_e and HTO connected porosity ε_c using diffusion cells [1].

Sorption experiments (²²⁶Ra):

Batch sorption experiments with different samples representative of the heterogeneity of the drill cores, S/L ratio = 117 g·L⁻¹, ²²⁶Ra analysis by liquid scintillation counting (LSC).

Modelling sorption data (bottom-up approach):

Phreeqc 3.6 [2] assuming that illite and illite/smectite mixed layers are the main sorbing phases in the sandy OPA (cf. [3]).

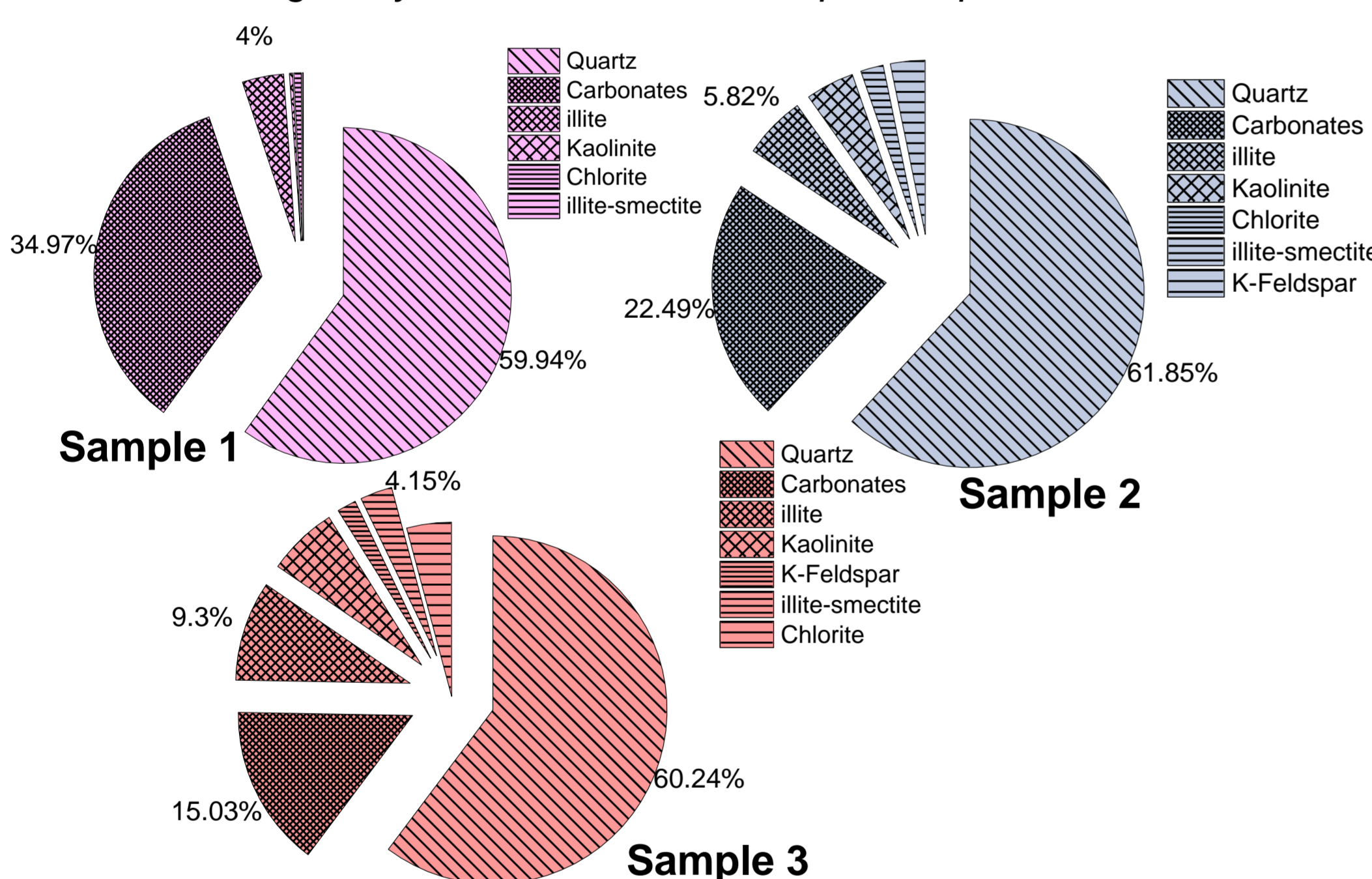
Blind reactive transport modelling of ²²⁶Ra migration in sandy OPA

- ICP 1.6 [4], Phreeqc 3.6 [2], Comsol Multiphysics 5.5 [5],
- Thermochemie database V10a,
- Retention of ²²⁶Ra is taken into account via cation exchange, surface complexation reactions and the formation of solid solutions of (Ra,Ba,Sr)-sulphates.

Selected results

Characterization of sandy OPA

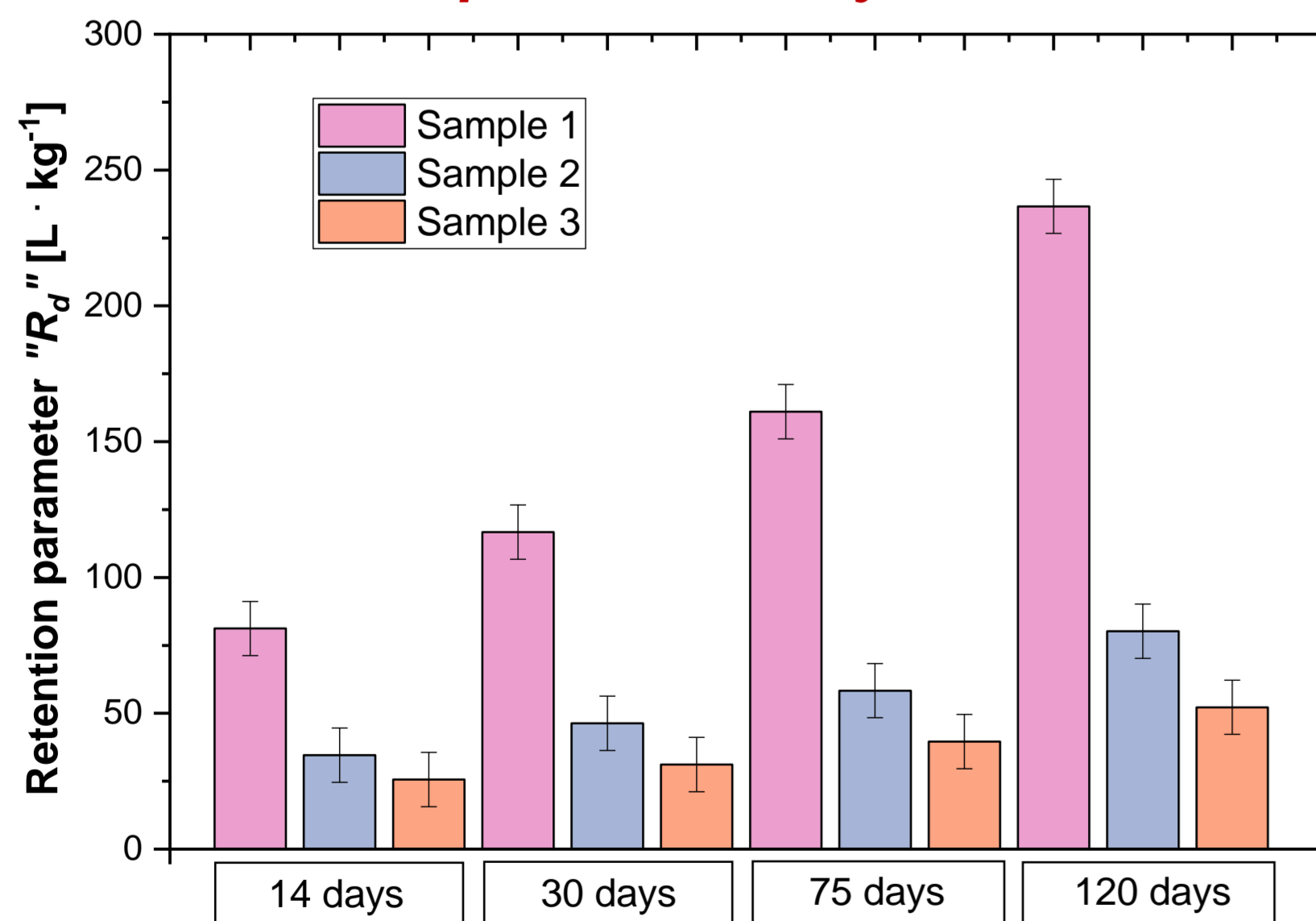
Mineralogical composition (wt.%) of powdered samples representative of the heterogeneity of the core used in sorption experiments



→ Minor phases: pyrite, siderite, feldspar, and rutile

→ (Ba, Sr) sulphate solid solutions embedded in clay matrix

²²⁶Ra sorption on sandy OPA



Duration of the experiments

Sorption of ²²⁶Ra on samples selected from sandy OPA facies as a function of time (S/L ratio = 117 g L⁻¹; $c_0(\text{Ra}) \sim 10^{-9}$ mol·L⁻¹).

→ $20 \text{ L} \cdot \text{kg}^{-1} < R_d(^{226}\text{Ra}) < 125 \text{ L} \cdot \text{kg}^{-1}$ (after 30 days)

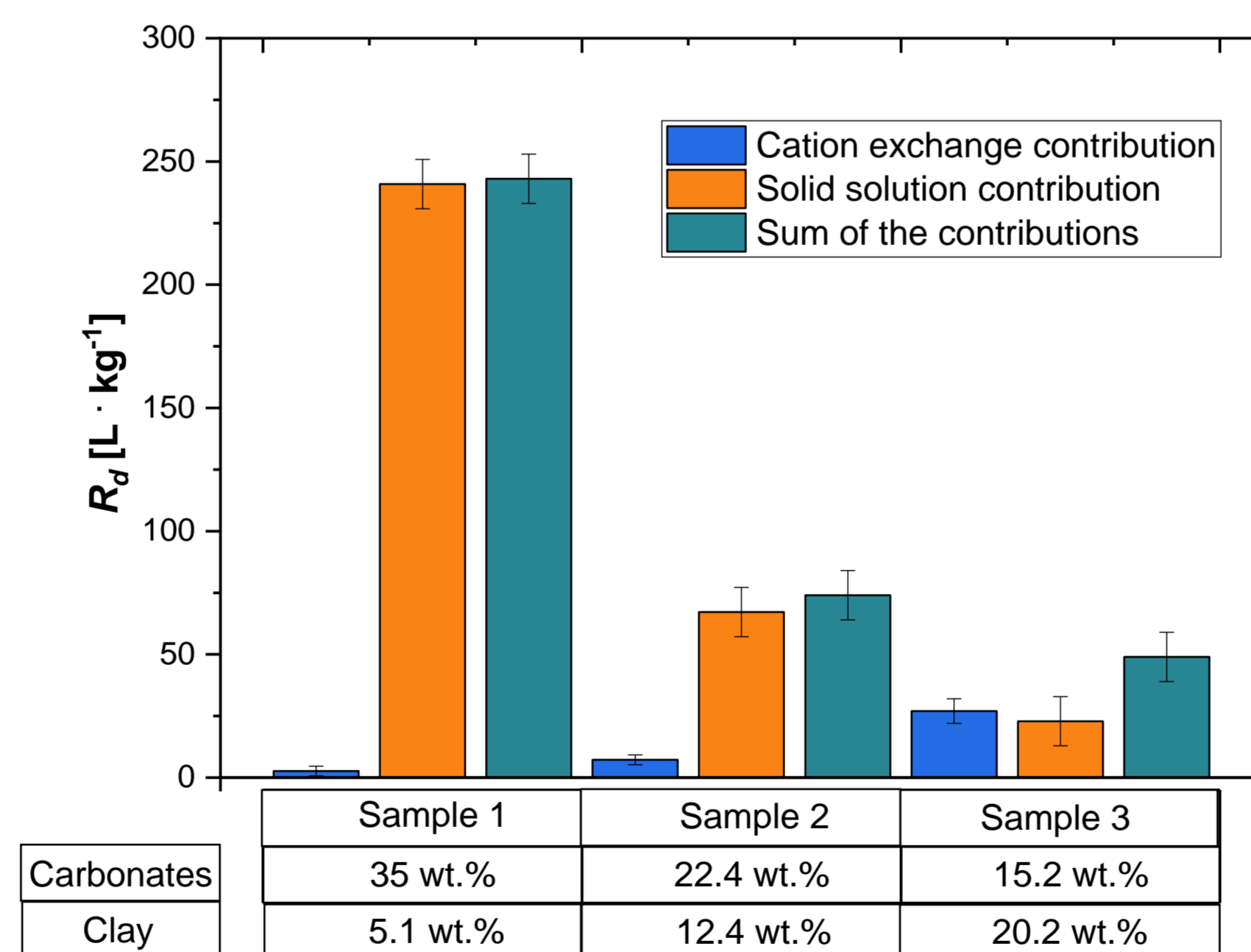
→ Increase of ²²⁶Ra retention over time and with increasing carbonate content

→ Diffusion or incorporation of ²²⁶Ra into minerals (e.g., calcite) indicated

²²⁶Ra sorption modelling (bottom-up approach)

Surface complexation reaction	Reference
$\equiv \text{clay_OH} + \text{Ra}^{2+} = \equiv \text{clay_ORa}^{+} + \text{H}^{+}$	[7]
Cation exchange reaction	
$2\text{Na}^{+} \text{clay} + \text{Ra}^{2+} = \text{Ra_clay} + 2\text{Na}^{+}$	[7]

- Site types, site capacities, and protolysis constants for illite [6]
- Parameters for ²²⁶Ra sorption on pure clay phases [7]



Different contributions to ²²⁶Ra retention in sandy OPA after 120 days equilibration time

→ The results show a major contribution on ²²⁶Ra retention due to formation of Ba–Sr–Ra carbonates and sulfate solid solutions and a smaller contribution due to surface complexation and cation exchange.

Prediction of ²²⁶Ra migration in sandy OPA

- Site types, site capacities, and protolysis constants for illite [6]
- Parameters for ²²⁶Ra sorption on pure clay phases [7]
- Clay water saturated with calcite, dolomite, celestine and barite
- Boundary conditions: left → fixed concentration, right → closed

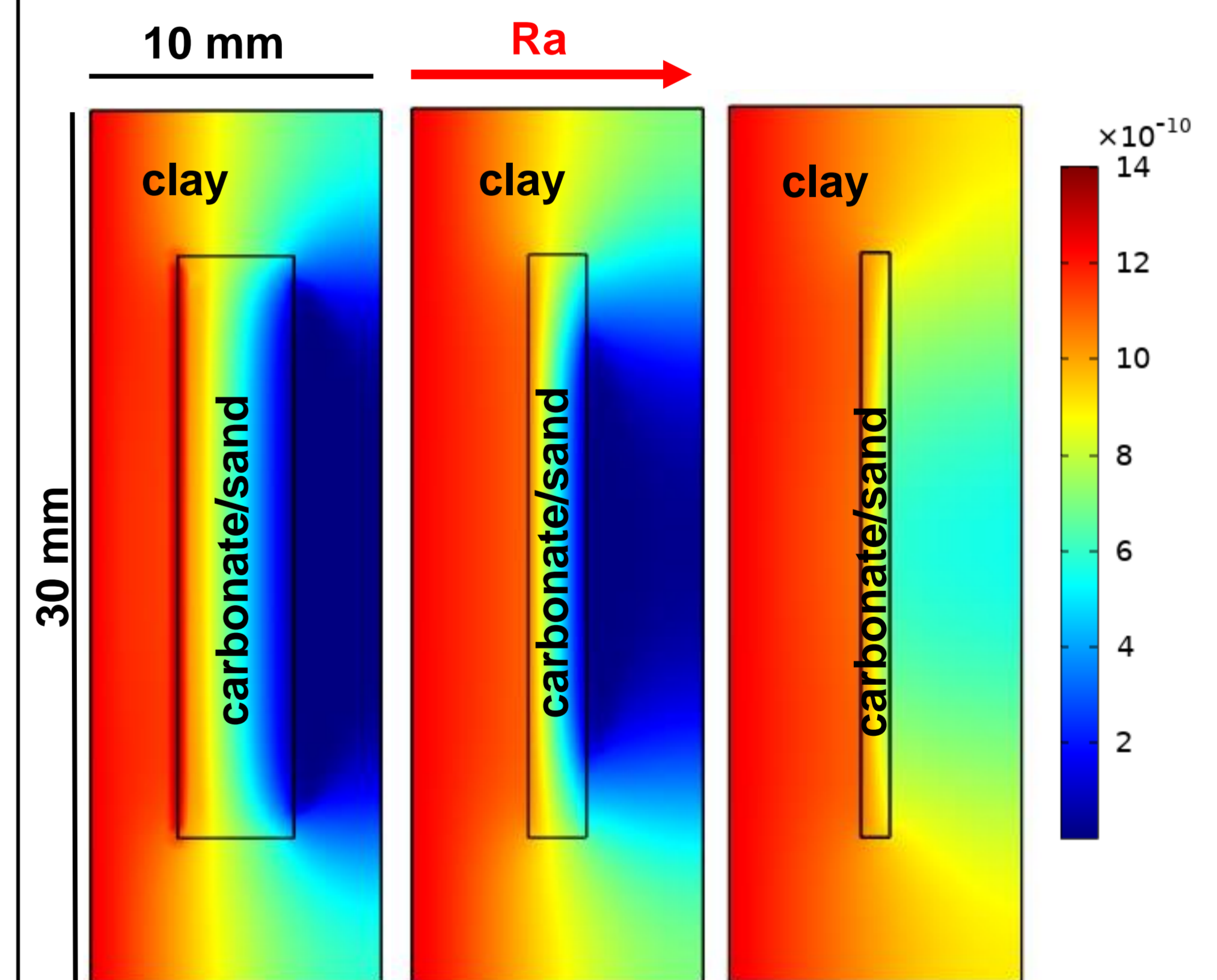
Clay layer:

- CEC = $106 \cdot 10^{-3}$ eq kg⁻¹ OPA
- $D_e = 4 \cdot 10^{-11}$ m²/s, porosity $\varepsilon = 0.16$ [8]

- (Ra,Ba,Sr)-sulphate solid solution formation in equilibrium with clay water

Carbonate/sand lens:

- Porosity $\varepsilon = 0.08$ [9]
- $D_e = 2 \cdot 10^{-12}$ m²/s
- Different proportions of clay and carbonate/sand



Modelled diffusion profiles of ²²⁶Ra (mole/kg_{water}) in sandy OPA after 10 days of in-diffusion.

→ The reactive transport model shows different diffusion profiles of ²²⁶Ra as a function of carbonate/sand content

Conclusions

Characterization of sandy OPA samples

→ Heterogeneous composition with amounts of quartz, carbonates, and clay minerals varying from 58 to 65 wt.%, 13 to 35 wt.% and 5 to 25 wt.%, respectively.

Sorption parameters

→ Increase of ²²⁶Ra retention over time indicating

→ kinetically controlled sorption process.

→ Uptake of ²²⁶Ra associated with cation exchange, surface complexation reactions, and

→ Formation of solid solutions of ²²⁶Ra with (Ba, Sr)-sulphates and/or uptake by carbonate minerals.

→ Bottom-up approach for sorption modelling not applicable for ²²⁶Ra in clay rocks with high carbonate content.

Reactive transport model of Ra migration

→ The heterogeneity of OPA-SF affects the diffusive mass transport of ²²⁶Ra.