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CENTRE D'ÉTUDE DE L'ÉNERGIE NUCLÉAIRE

Diffusion of cementitious pore fluids into Boom Clay from a deep HLW disposal site: modeling of a laboratory experiment and long term interaction

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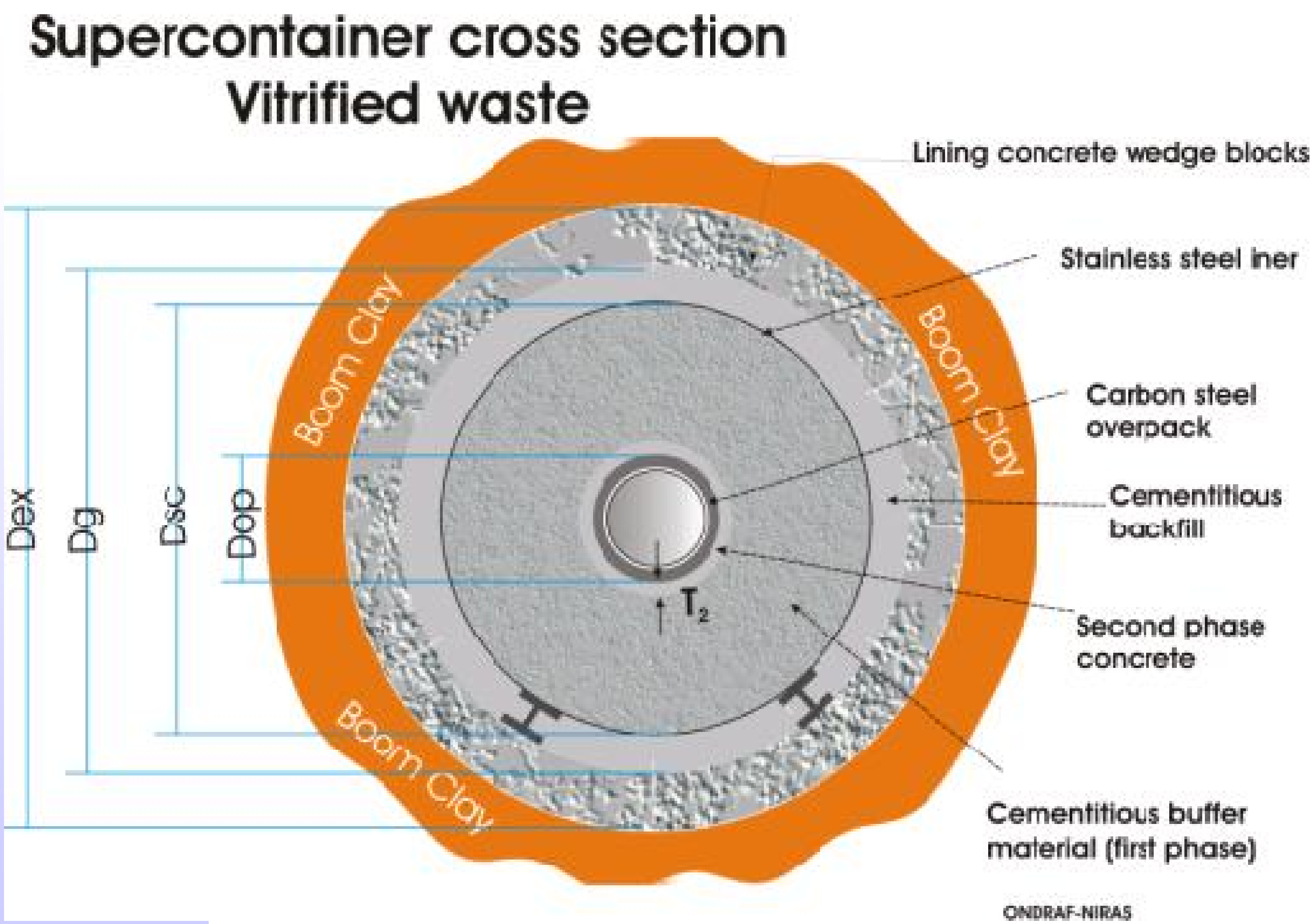
Outline

- Background
- Objective
- Calibration and modelling of laboratory experiments with young concrete water
- Long-term diffusion modelling
- Conclusion

Engineered barrier system HLW in Boom Clay



Objective

- To model laboratory experiments of advection of young concrete water throug Boom Clay cores
- Assess the possible extent of Boom Clay alterations owing interactions with alkaline fluids for a period of 25000 years

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Calibration and modelling of laboratory experiments with young concrete water

Experimental set-up

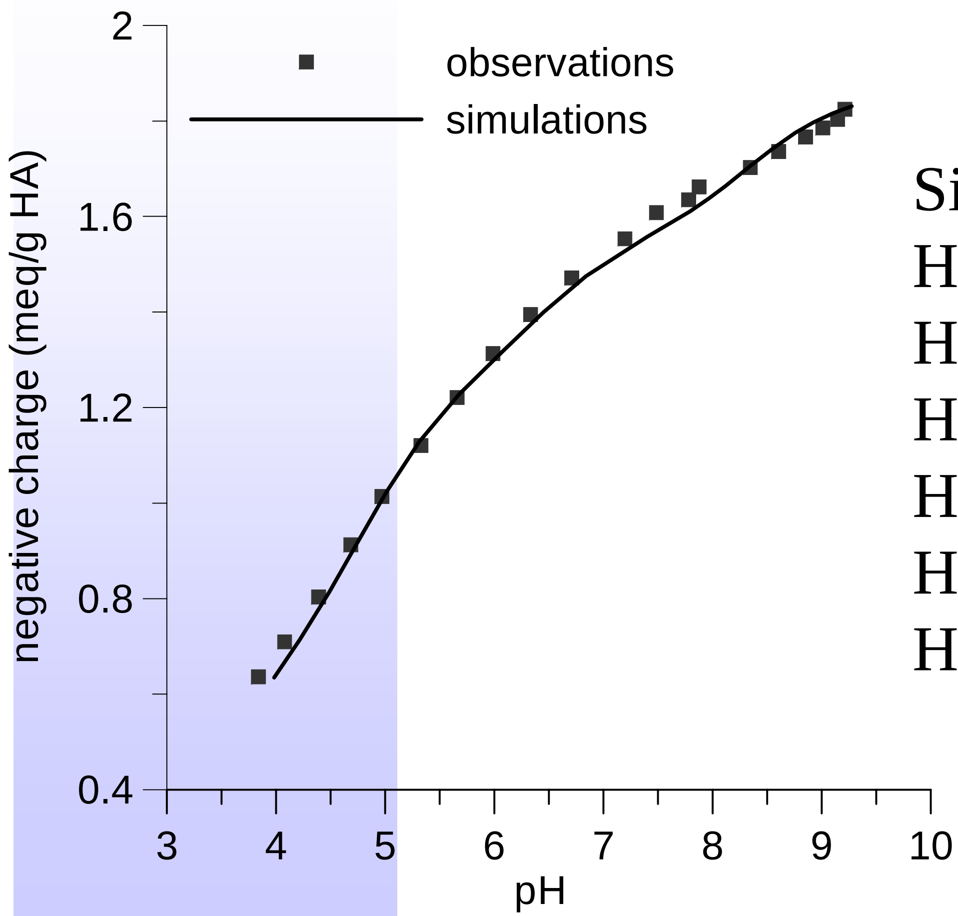
- Boom Clay core (32 mm)
- Inflow with young concrete water
  - pH 13.1
  - High K and Na content
- Steady-state flow conditions for 1000 d
- Measuring pH, Na, K, Si, Al, Ca, Mg in the outflow

Boom clay model

- Primary minerals
  - Quartz, kaolinite, illite, Na-montmorillonite, calcite
  - Kinetic dissolution/precipitation reactions for first 4:
    - Based on transition state theory
    - pH-dependent
    - Uncertain parameter: reactive surface area
- Solid – solution reactions
  - Ion exchange on fixed CEC-complex (clay)
  - Ion exchange on pH-dependent CEC complex (organic matter)
  - Proton surface complexation on illite/montmorillonite

Solid – solution reactions

- pH-dependent cation exchange complex



Site	pKa	size(meq/g om)
HYa	1.65	0.628
HYb	3.3	0.628
HYc	4.95	0.300
HYd	6.85	0.300
HYe	9.6	0.300
HYf	12.35	0.300

Solid – solution reactions

- pH-dependent cation exchange complex
- Proton surface complexation (Bradbury et al., 2005)
  - 3 surface complexation sites (2 weak and 1 strong site)
  - Related to illite and montmorillonite
- Equilibrium reactions for secondary phases

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Model approach

- Three types of models
  - Model 1: fixed CEC
  - Model 2: fixed CEC + pH dependent CEC
  - Model 3: fixed + pH-dependent CEC + SC
- Calibration of uncertain parameters
  - RSA of primary minerals (4)
  - Fixed CEC
  - Total size of pH-dependent CEC
  - Total size of surface complexation sites
  - (choice of secondary minerals)
- Reactive transport model: PHREEQC

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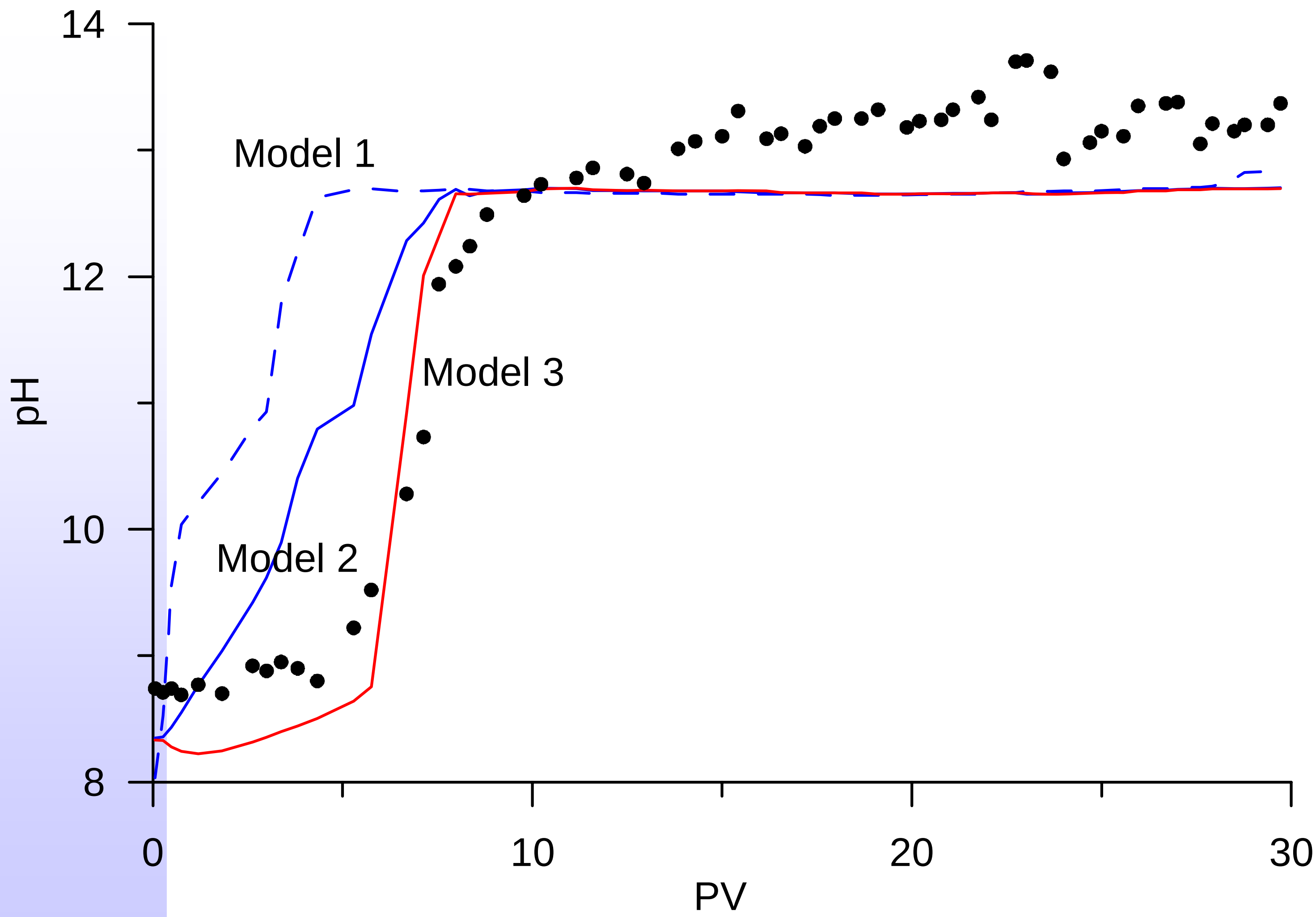
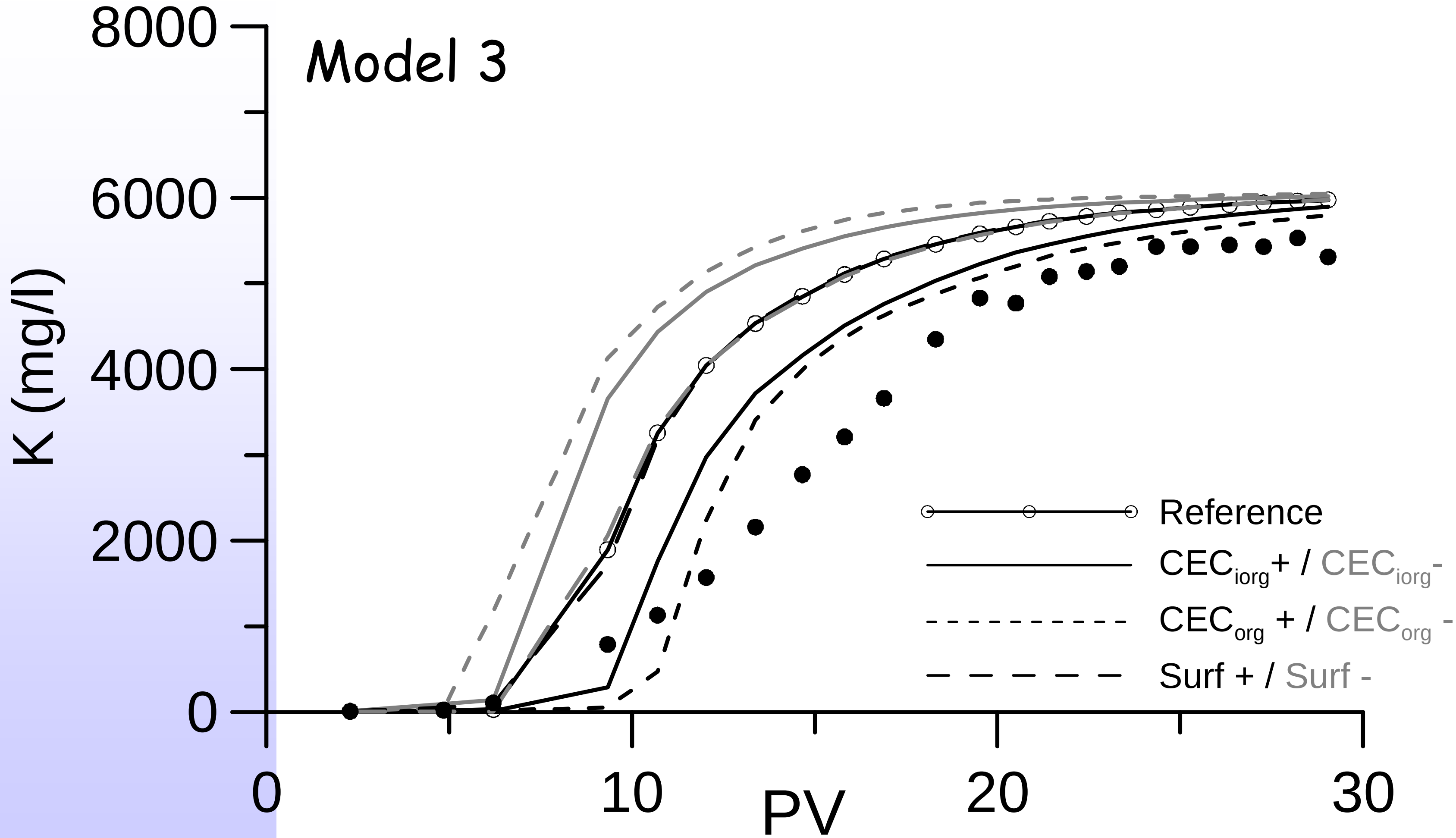
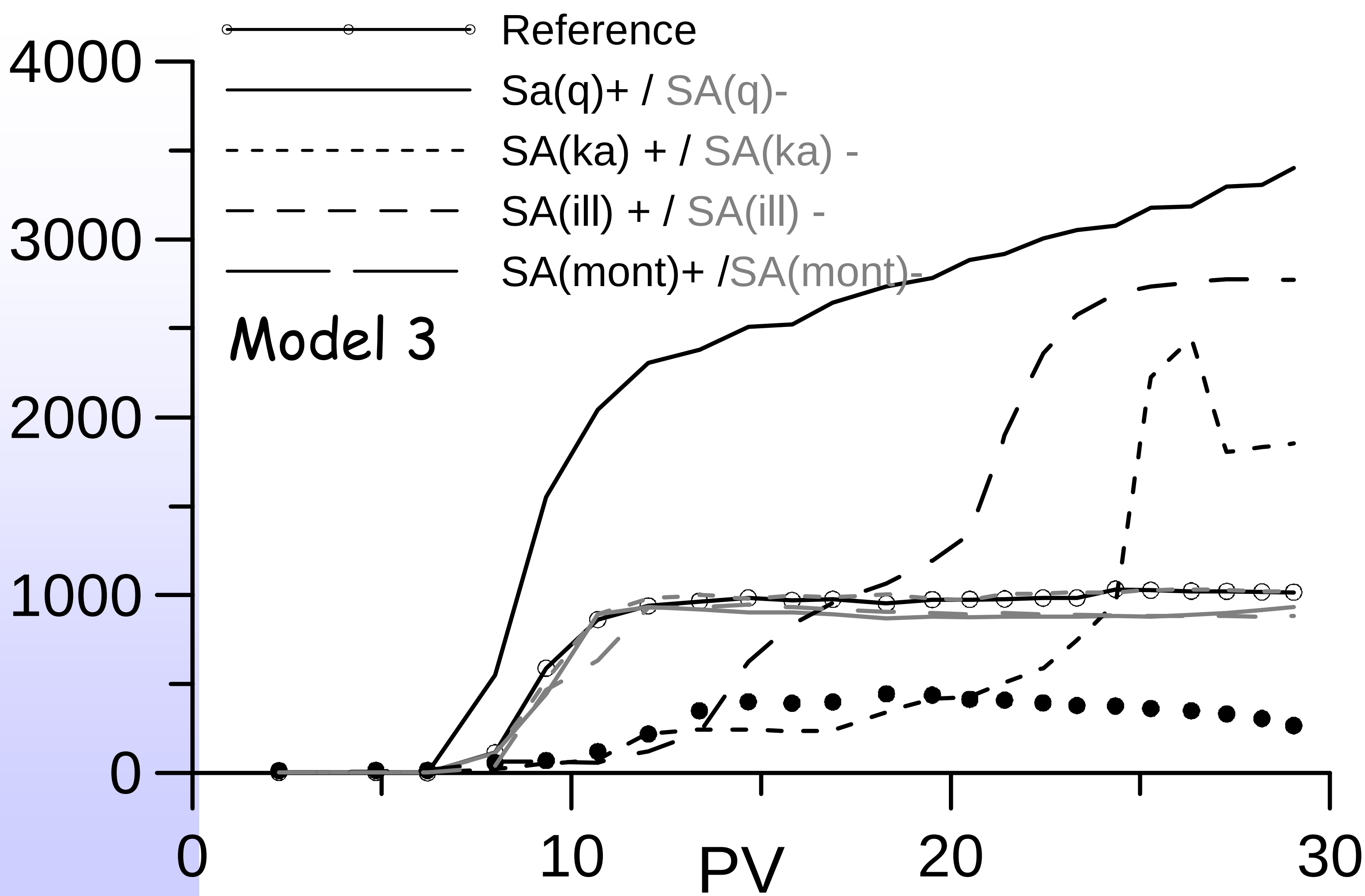
Model comparison simulation with reference parameters



Sensitivity of parameters Na, K, and pH to ion exchange and SC parameters

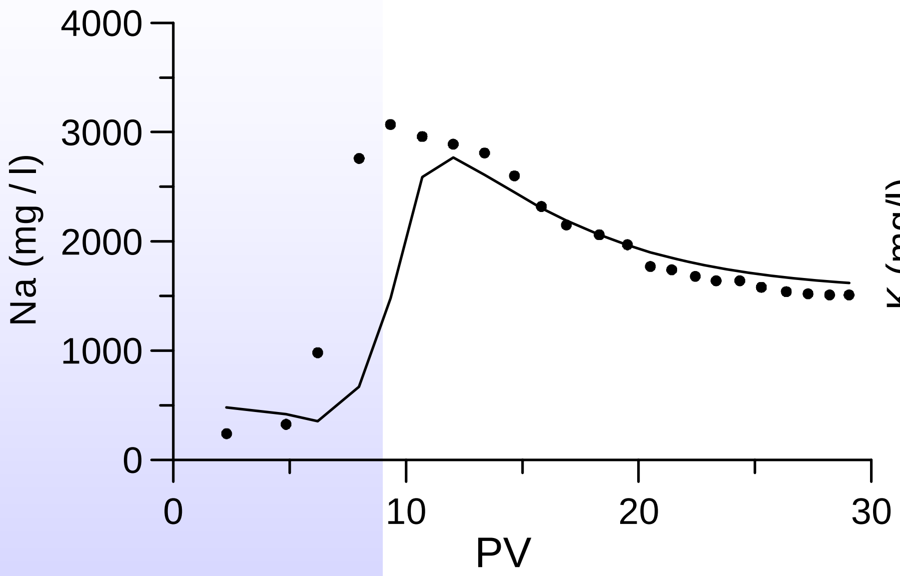


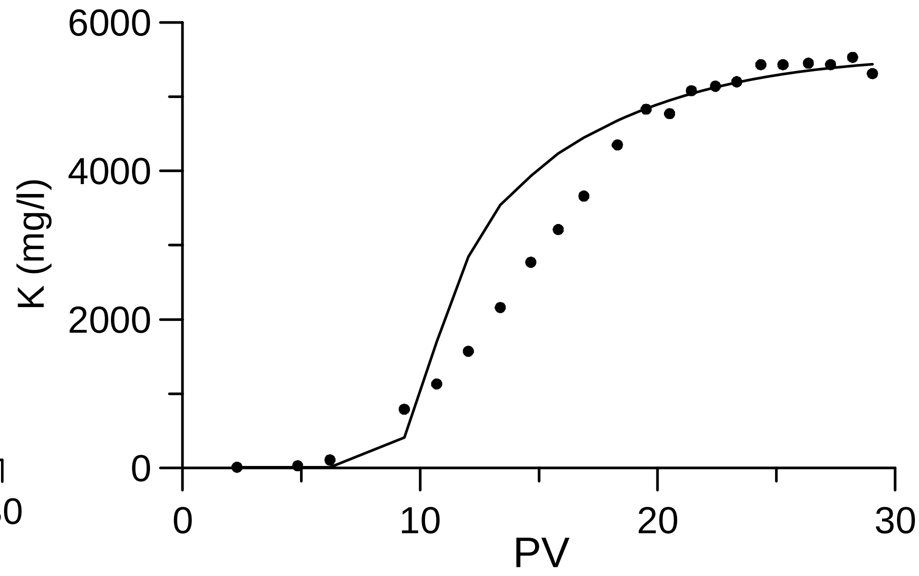
Sensitivity of parameters Al and Si to RSA



Calibration results

- Na and K: typical chromatographic behaviour

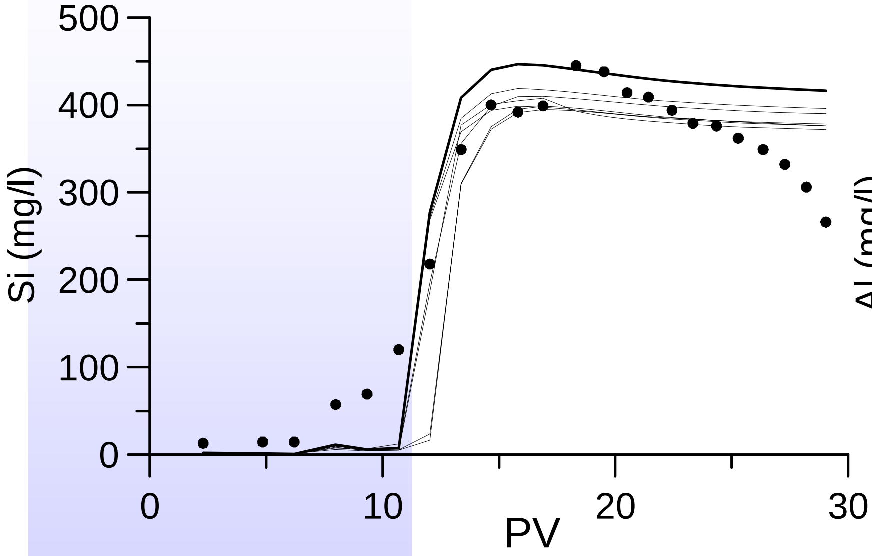


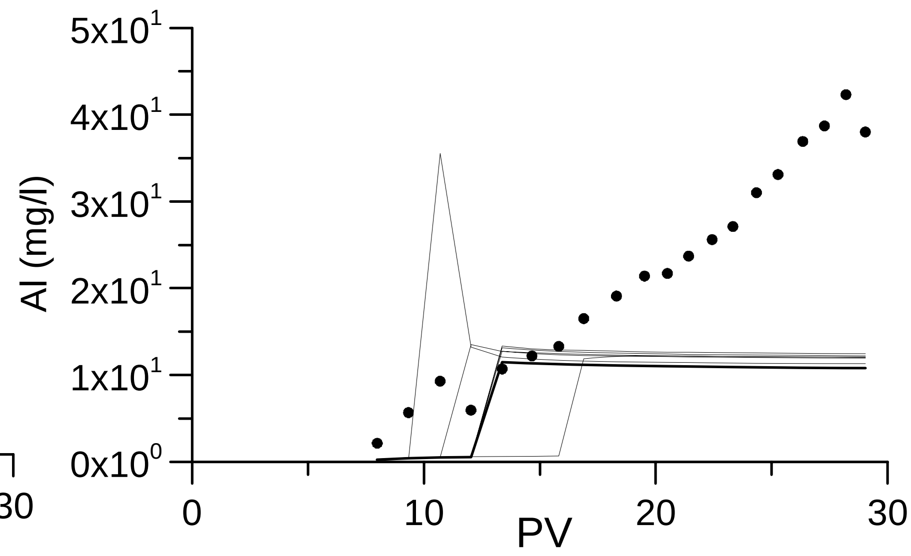


Model 3

Calibration results

- Al and Si: Kinetic dissolution primary minerals

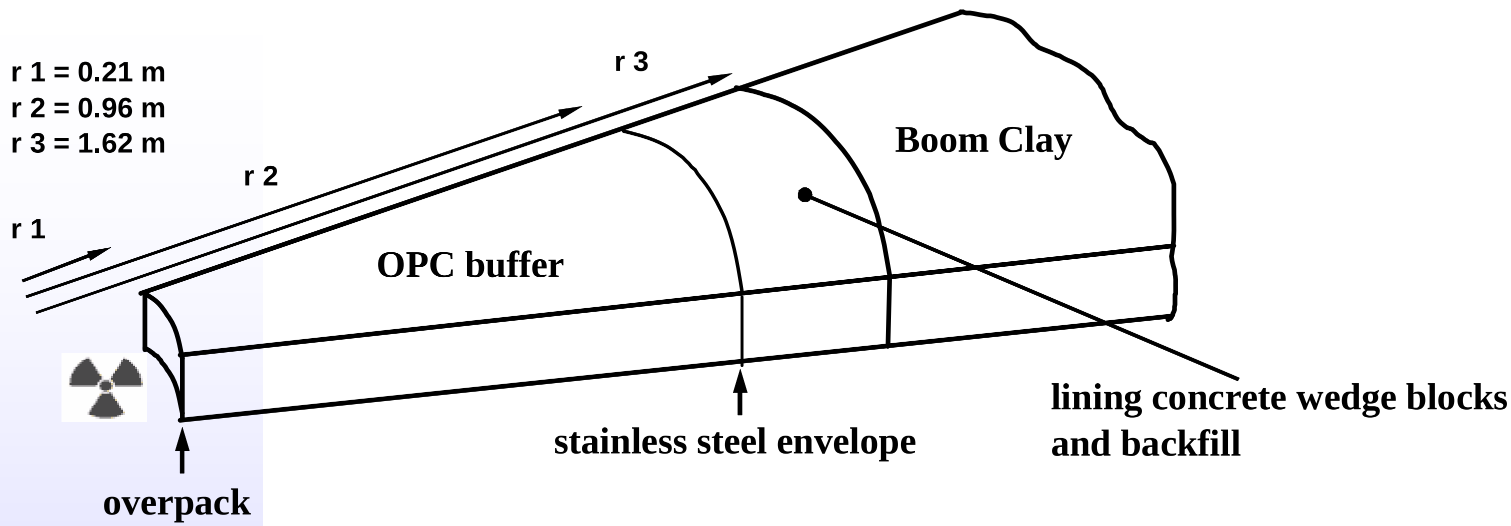




Model 3

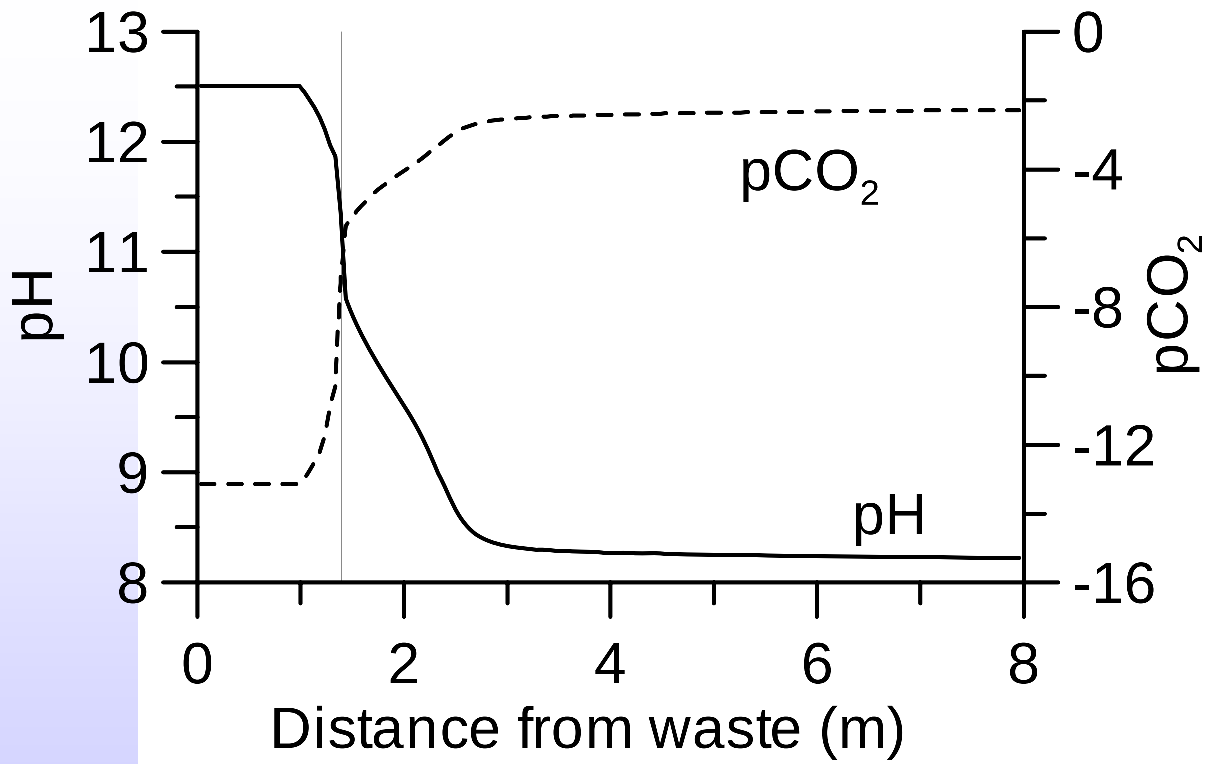
Long-term diffusion modelling

Model approach

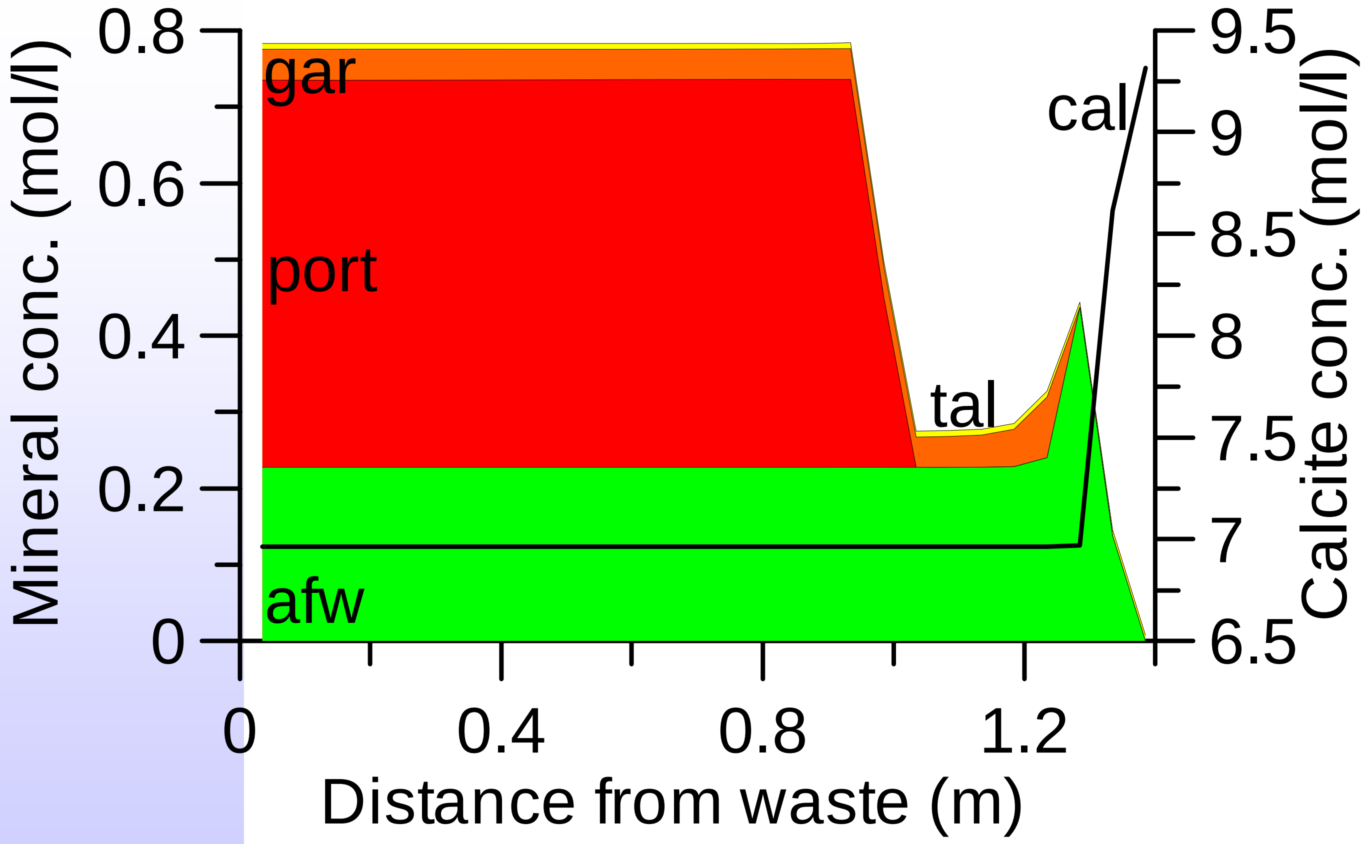


- Only equilibrium reactions
- Simplified concrete model (portlandite, afwillite, hydrogarnet, hydrotalcite, Na_2O, K_2O)
- Radial diffusion
- No feed-back from chemistry on porosity and D

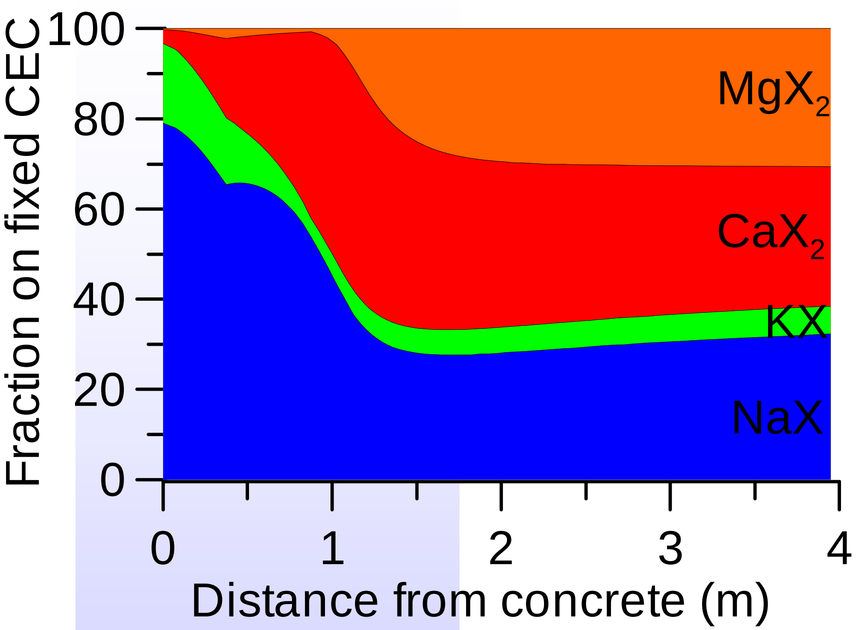
pH and PCO_2 after 25000 y

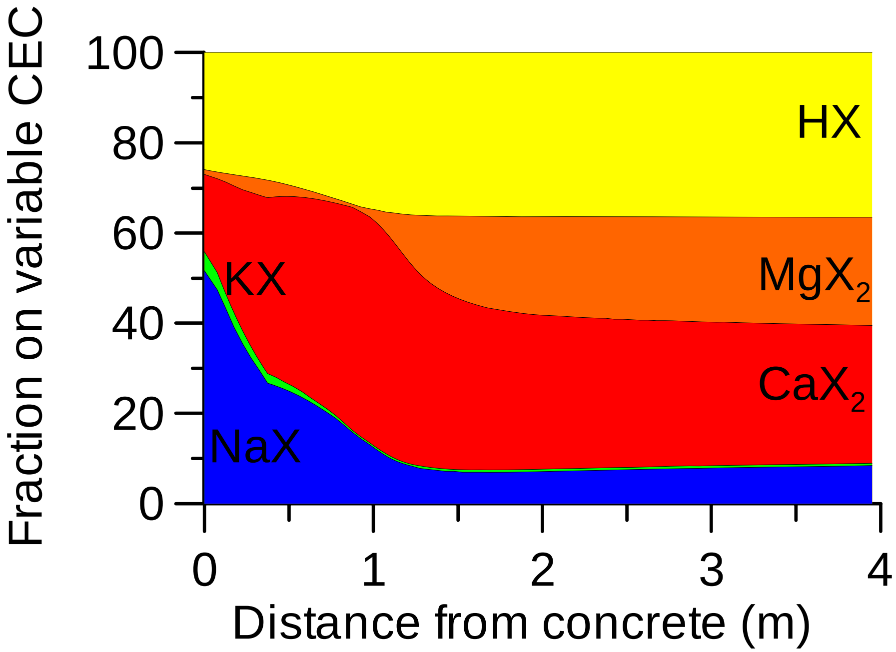


Changes in the concrete



Changes in the ion exchange complex





Conclusions

- Laboratory experiments
  - Exchange processes important
  - Kinetic dissolution of primary mineral (RSA uncertain parameter)
  - Secondary phases not so important
- Long term diffusion modelling
  - Equilibruim dissolution primary minerals
  - Further analysis: sensitivity of model assumptions
    - Concrete model
    - Choice secondary minerals
    - Boom clay model

- Acknowledgement
  - NIRAS/ONDRAF for financial support

- Contact
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