ECN

Energy research Centre of the Netherlands

Stabilised Waste - Cement barrier - Soil interfaces

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OVERVIEW

Problem definition

Release of non-interacting species (3 layer model)

Scale issues

Illustration of detailed chemistry

Chemistry of individual layers

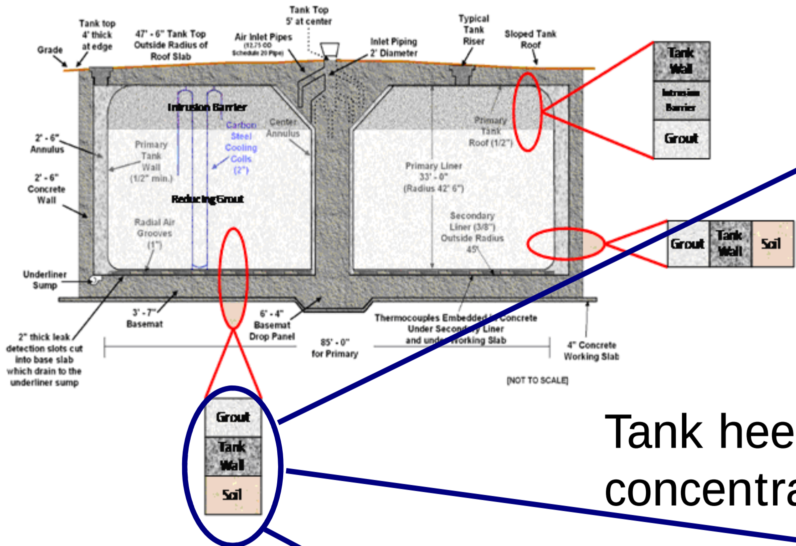
Interface reactions (diffusion in 3 layer model)

Carbonation and uncertainty

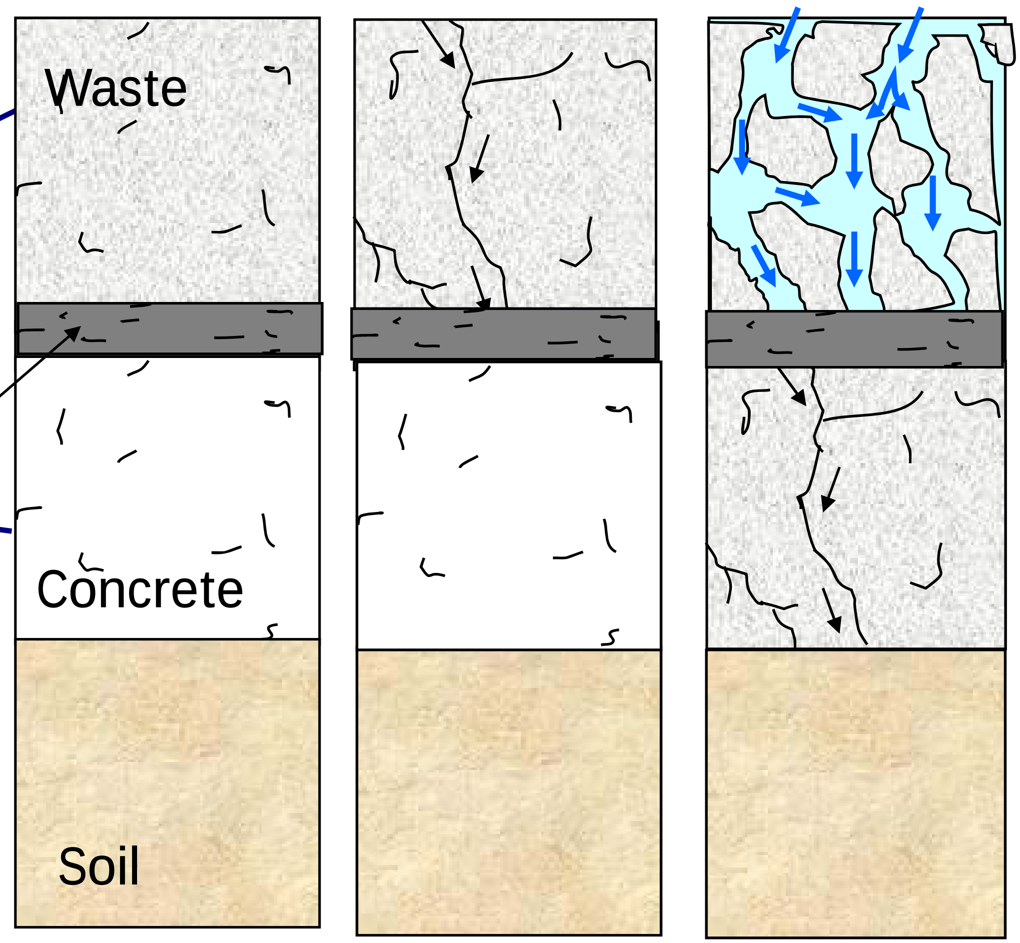
Conclusions

TANK CLOSURE

POSSIBLE CHANGES IN THE TANK CLOSURE SYSTEM AT LONG TERM

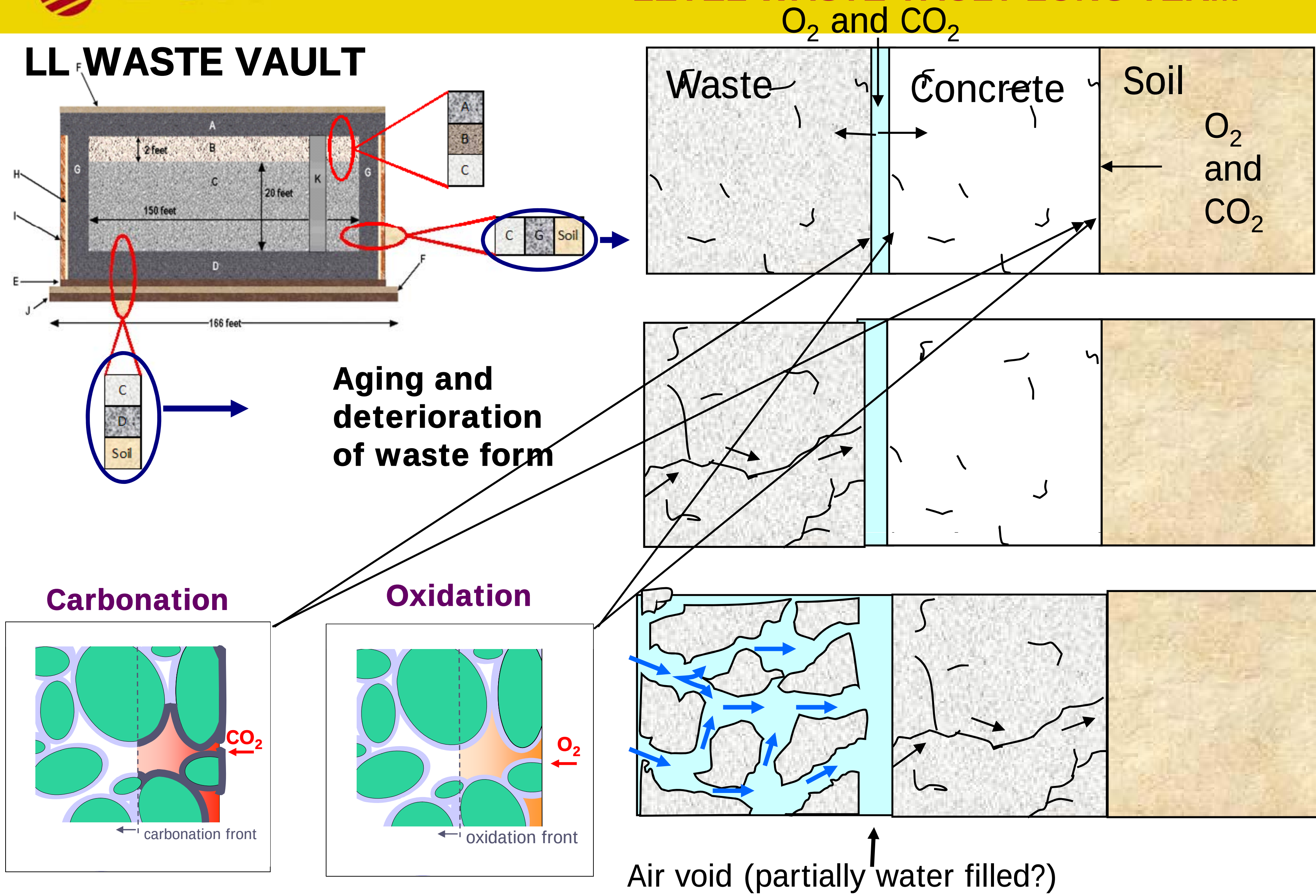


Tank heel concentrate



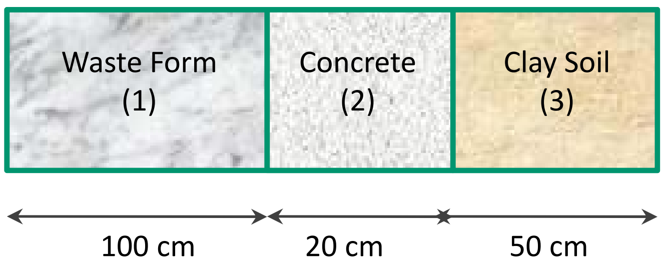
Aging and deterioration of waste form

POSSIBLE CHANGES IN THE LOW LEVEL WASTE VAULT LONG TERM



Effect of Boundary Conditions and Saturation on Release from a 3-layer System

3-Layer, 1-D diffusion model for non-interacting, conservative species (e.g., Na)



	Waste Form	Concrete	Clay Soil (compacted)
Density (g/cm^3)	1.7	2.4	1.8
Porosity	0.4	0.1	0.35
Tortuosity (sat'd)	5	15	2

<u>Boundary Conditions</u>

Waste form – no flux at interior boundary (left side)
Clay soil – zero concentration at external boundary (right side of clay soil)

<u>Initial Condition</u>

Na only in waste form at time zero at $C/C_0=1$

<u>Cases</u>

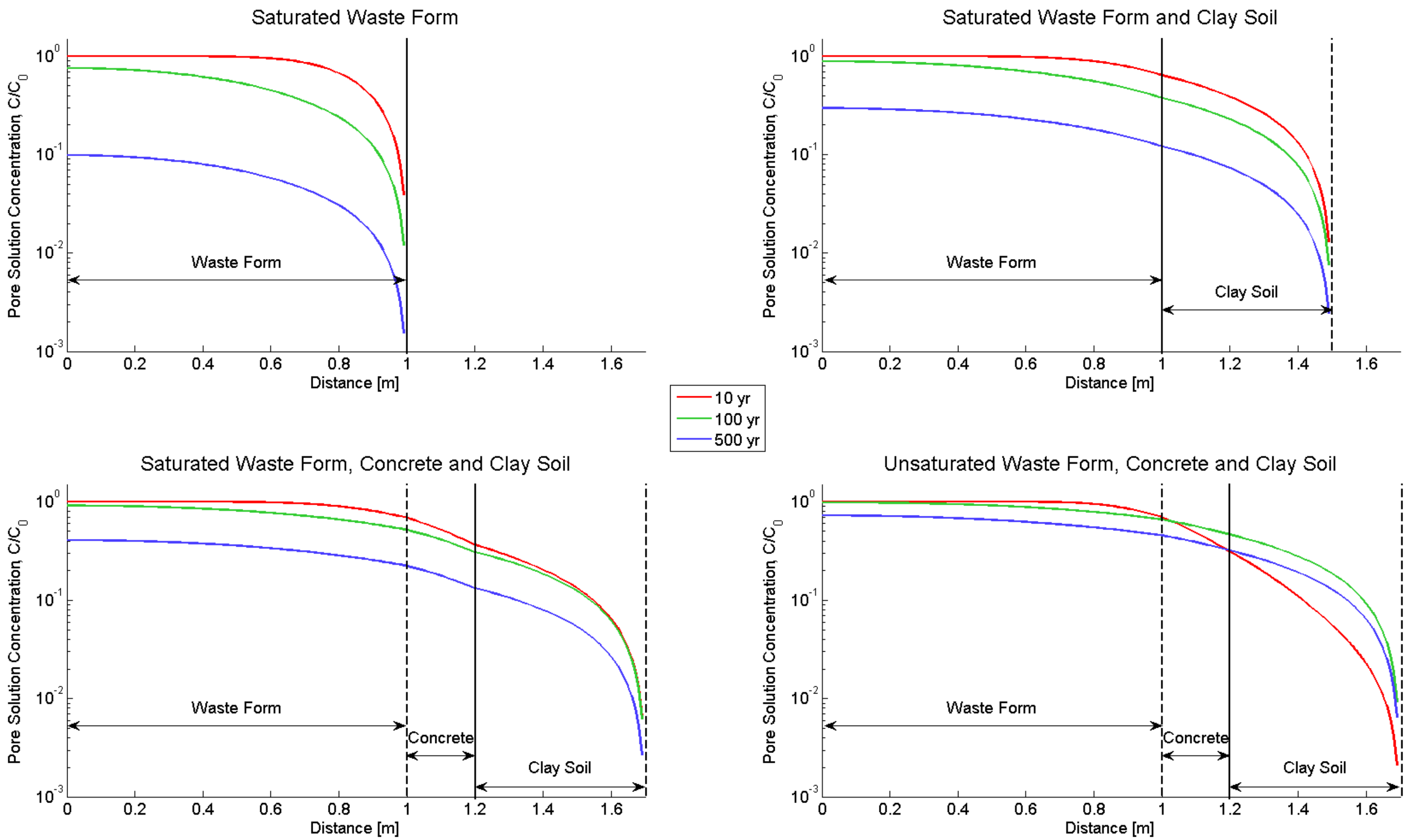
1. Saturated

2. Unsaturated (tortuosity values assumed 2x for both waste form and concrete and 3.2x for soil; available porosity assumed 0.8x for waste form and concrete and 0.16 x for soil)

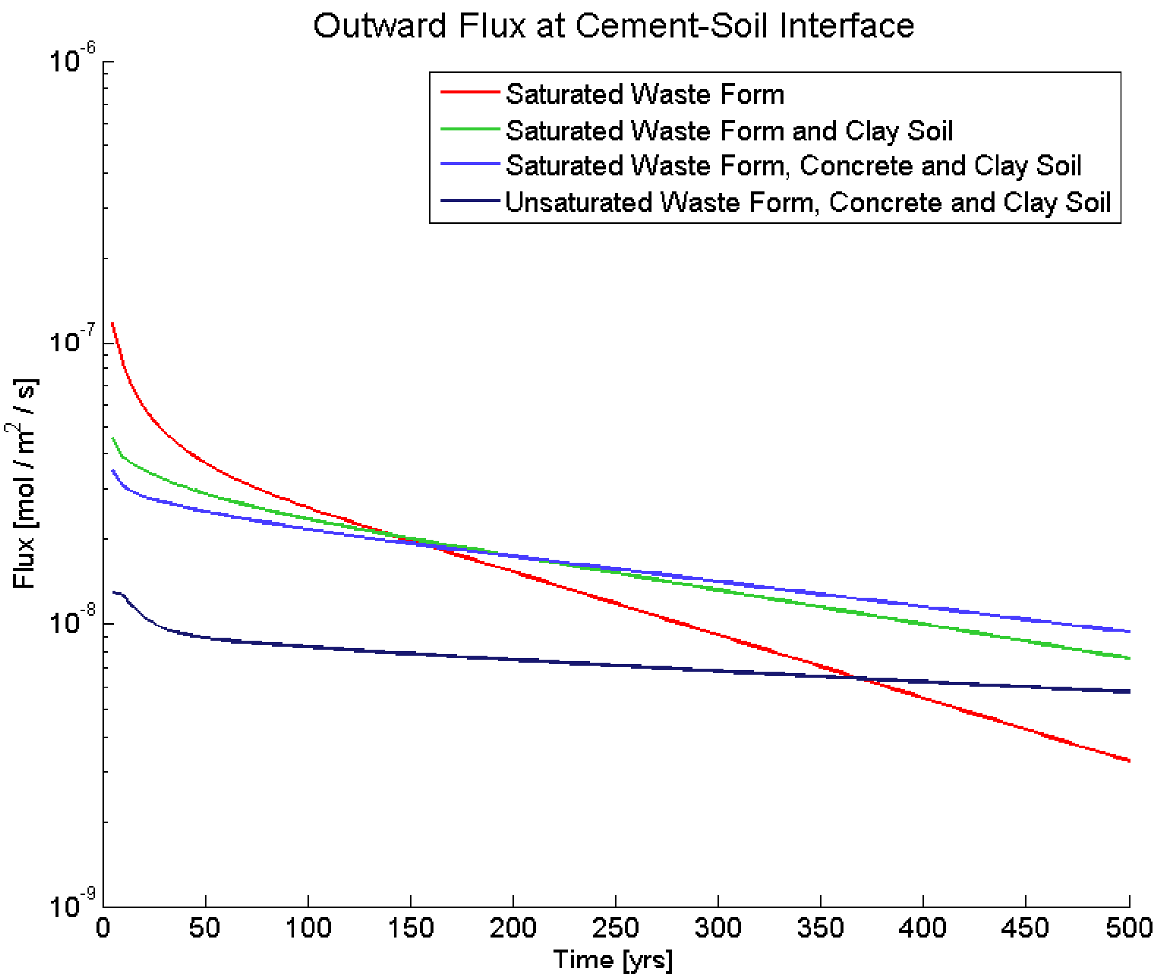
3. Same as (1) but with concrete layer only and $C/C_0=1$ at waste form-concrete interface and $C/C_0=0$ at concrete-clay soil interface

4. Same as (1) but with waste form only and $C/C_0=0$ at waste form boundary (waste form in infinite bath)

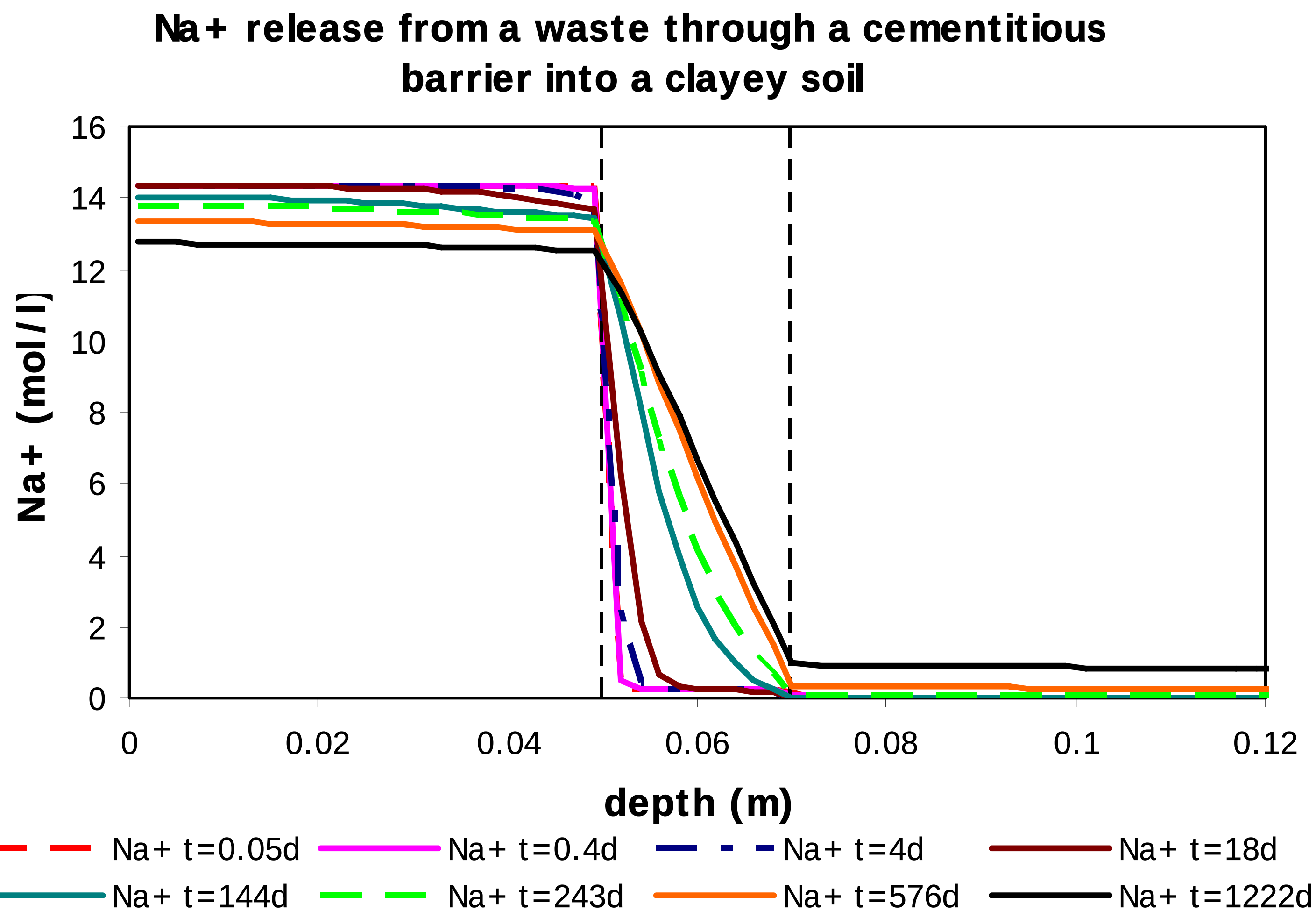
Transport of non-interacting species in 1-, 2-, and 3-layer system (sat – unsat)



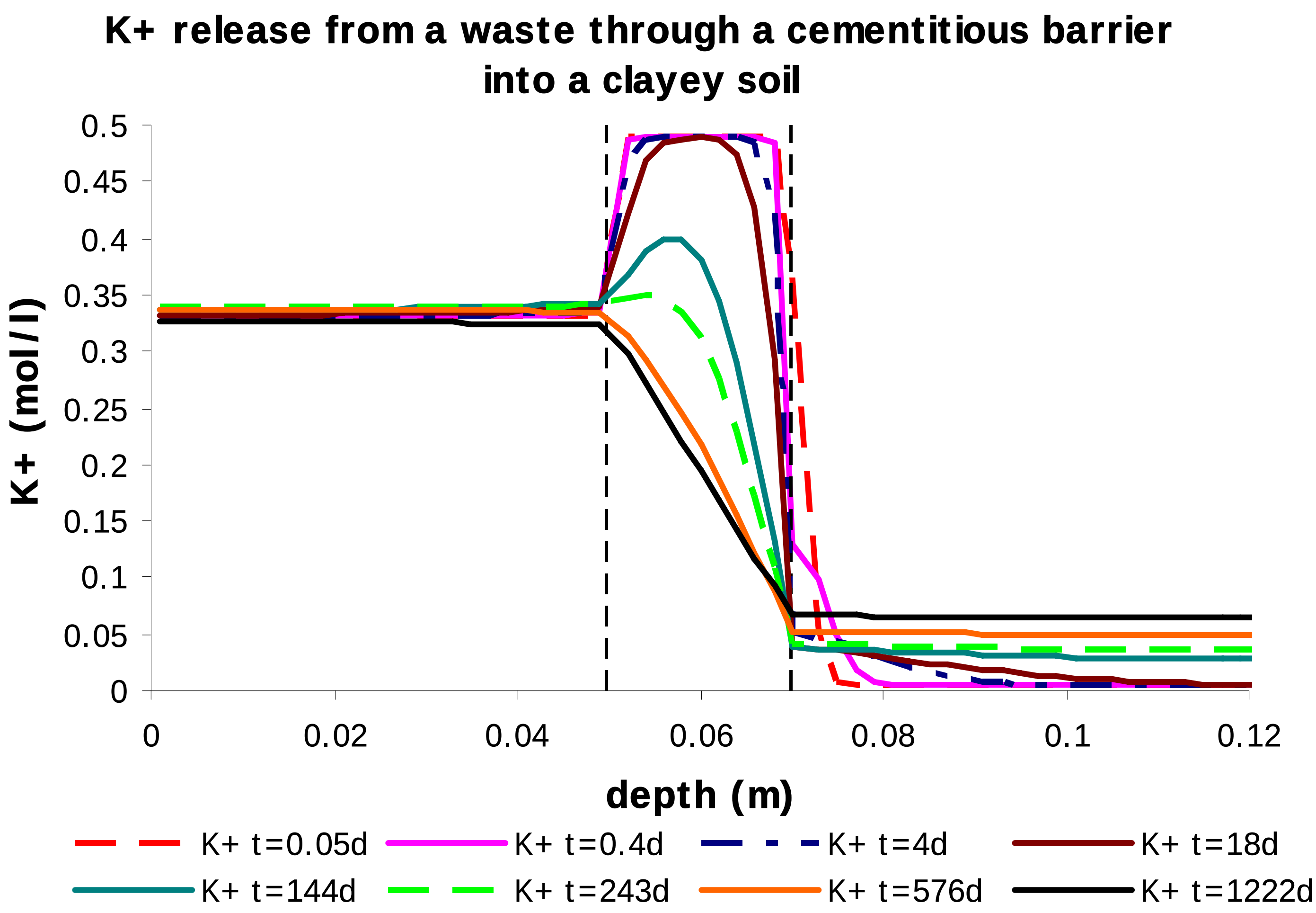
Flux of non-interacting species at the cement-soil interface



Scaling to smaller dimensions with same physical conditions



Steady state condition established within 2 years



In spite of higher K level in barrier steady state condition established within 2 years

Illustration of the importance of more detailed chemistry

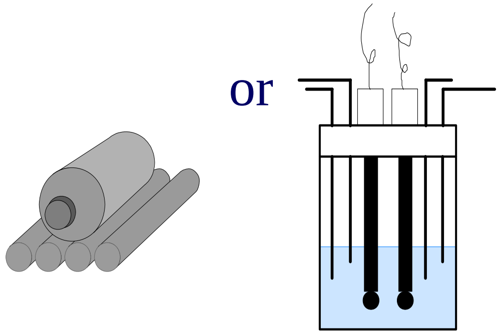
Geochemical speciation modeling based on pH dependence test results taking mineral precipitation, clay interaction, sorption on ironoxides, incorporation in ettringite and interaction with particulate and dissolved organic matter into account.

Sorption parameters for particulate and dissolved organic matter for U and Th based on the generic parameters derived by Milne et al, 2003 for the Nica Donnan model.

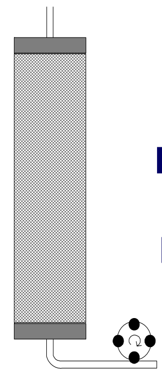
Characterisation leaching tests

Standardisation:
CEN/TC292, ISO/TC190,
CEN/TC345, CEN/TC351,
SW846

GRANULAR MATERIALS



pH DEPENDENCE TEST: BATCH MODE
ANC, CEN/TS 14429, or EPA method 1313 or, COMPUTER CONTROLLED CEN/TS 14997

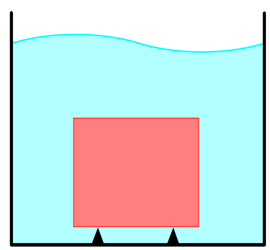


PERCOLATION LEACHING TEST
CEN TS 14405 or EPA method 1314

MONOLITHIC MATERIALS

Same as granular

+

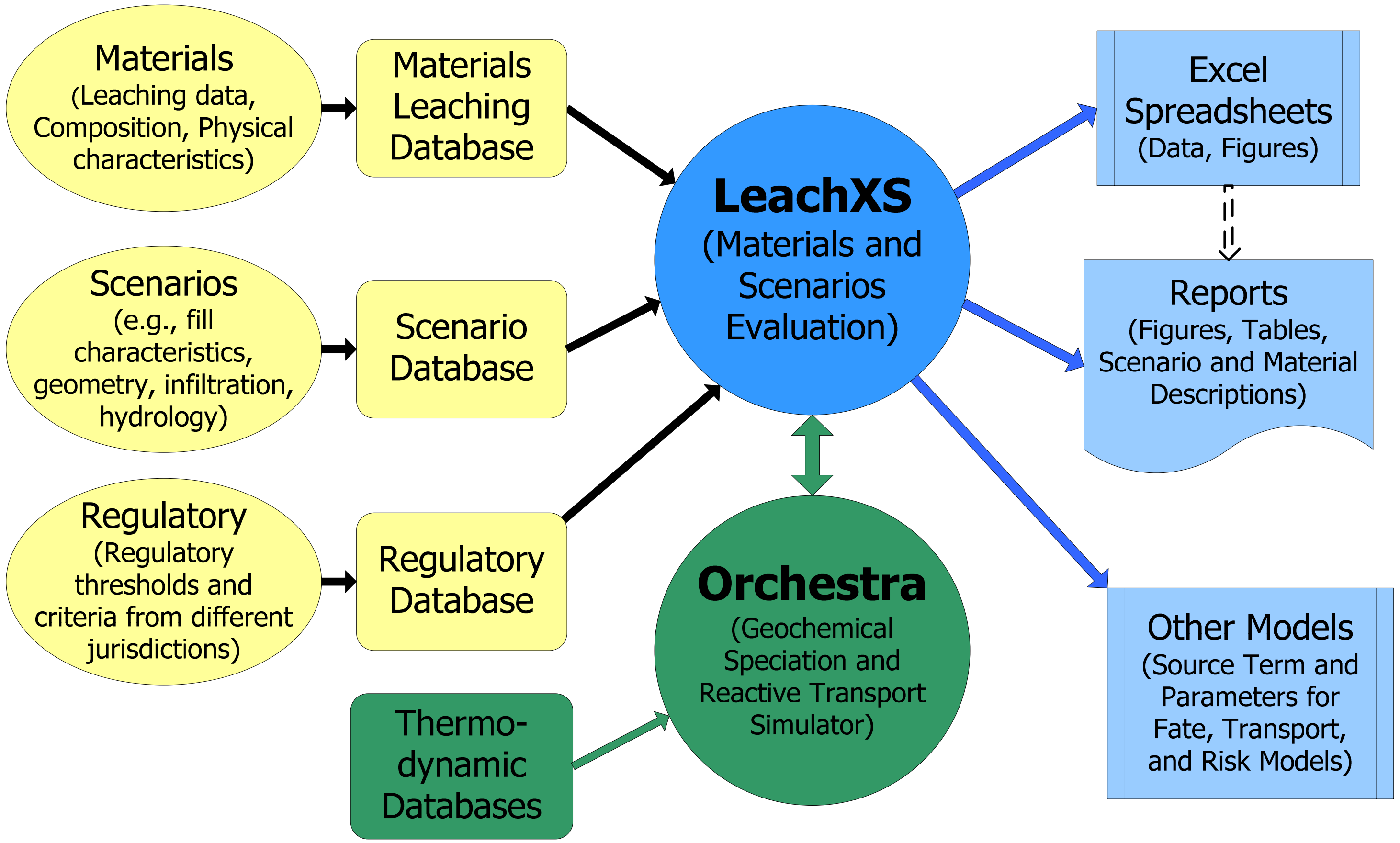


TANK LEACH TEST
MONOLITH CEN/TS 15863 and EPA method 1315 and COMPACTED GRANULAR LEACH TEST (NEN 7347 and EPA method 1313).

Chemical speciation aspects

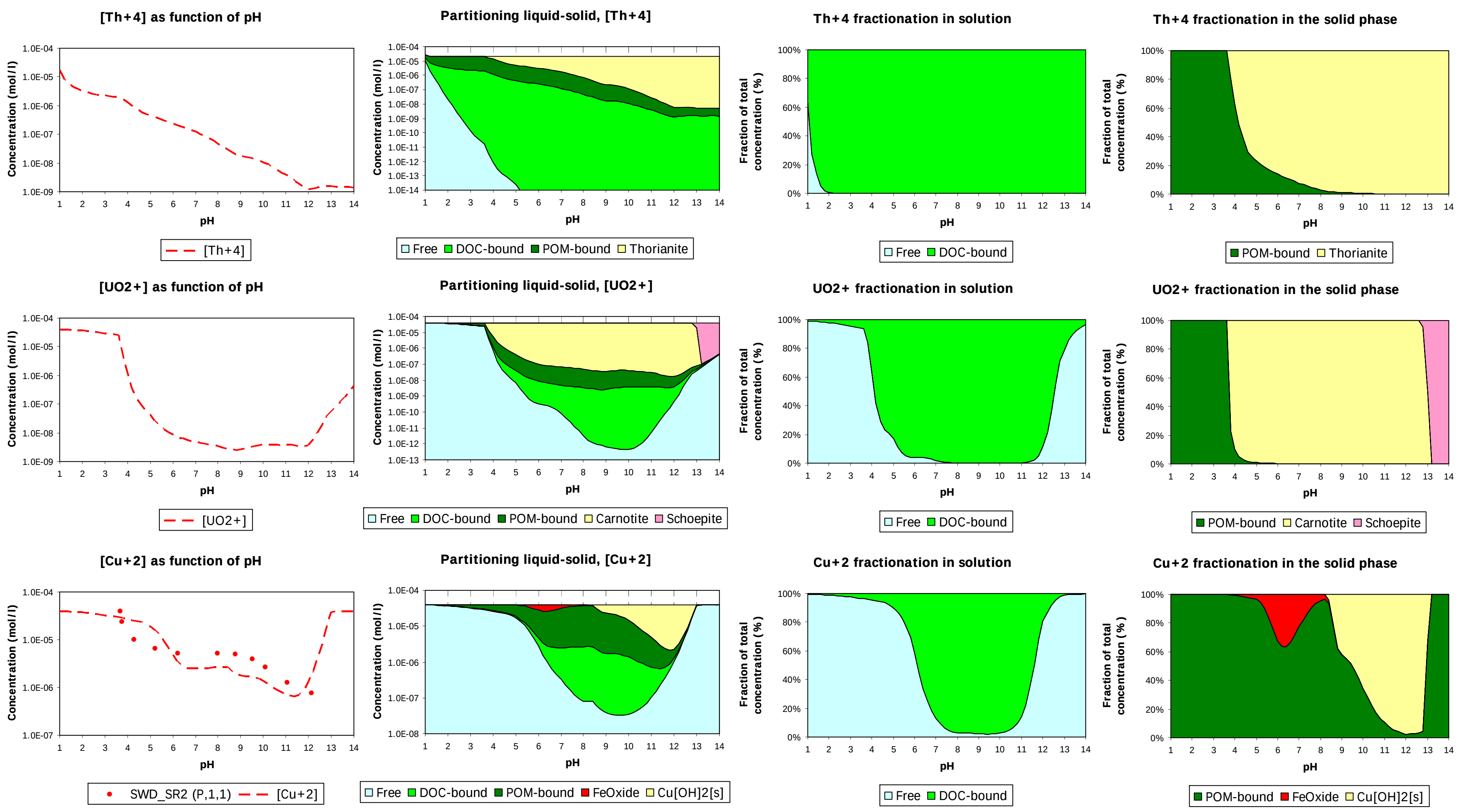
Time dependent aspects of release

LeachXS Structure



In the modeling mineral dissolution, sorption on hydrated ironoxides, clay interaction, interaction with particulate and dissolved organic matter and incorporation in ettringite solid solution.

Cement Stabilised Waste Mix



Information also relevant for stabilisation of contaminated soil

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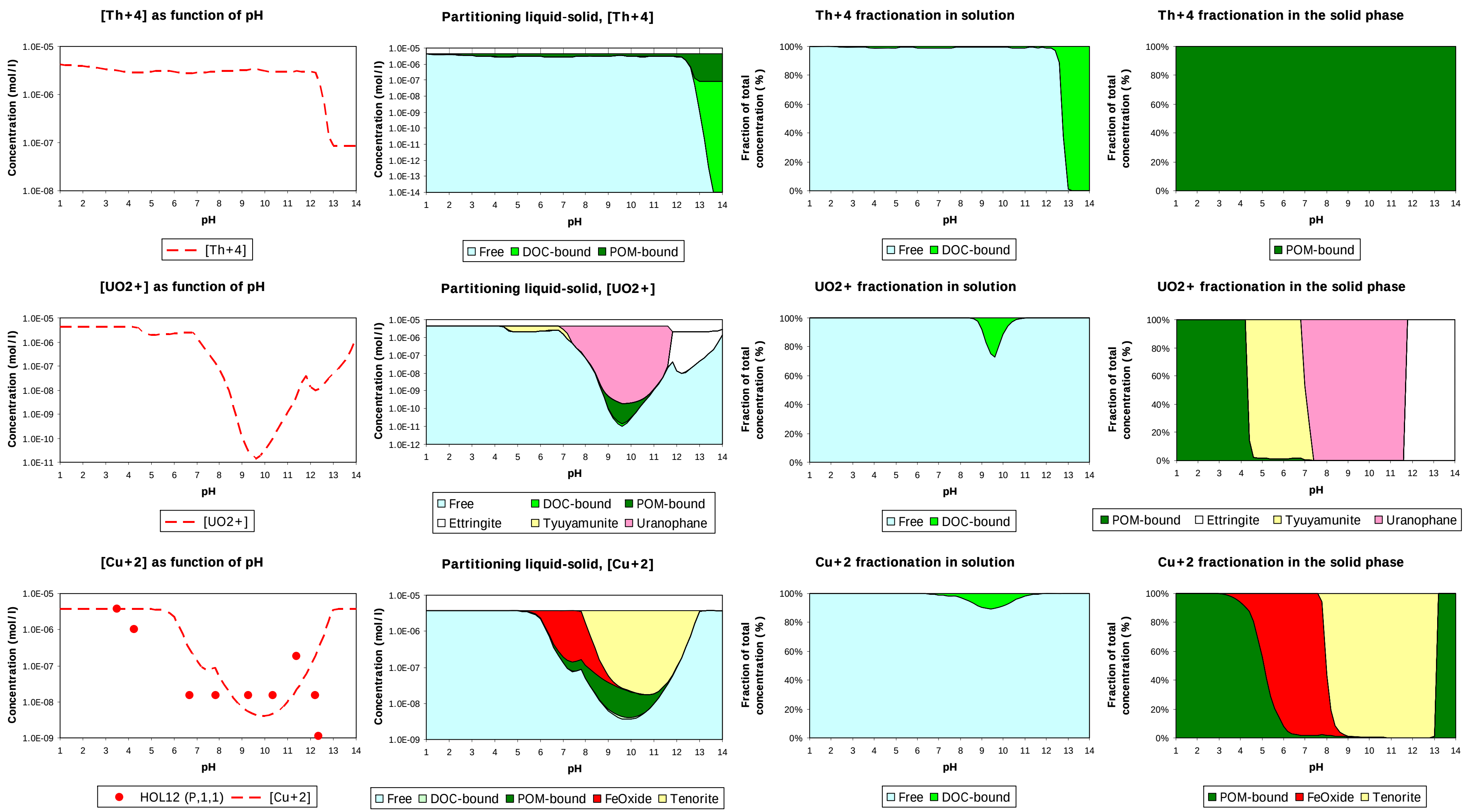
Granulated blast furnace slag – fly ash cement mortar



Cement mortars and concrete not inorganic: non-negligible organic matter content!

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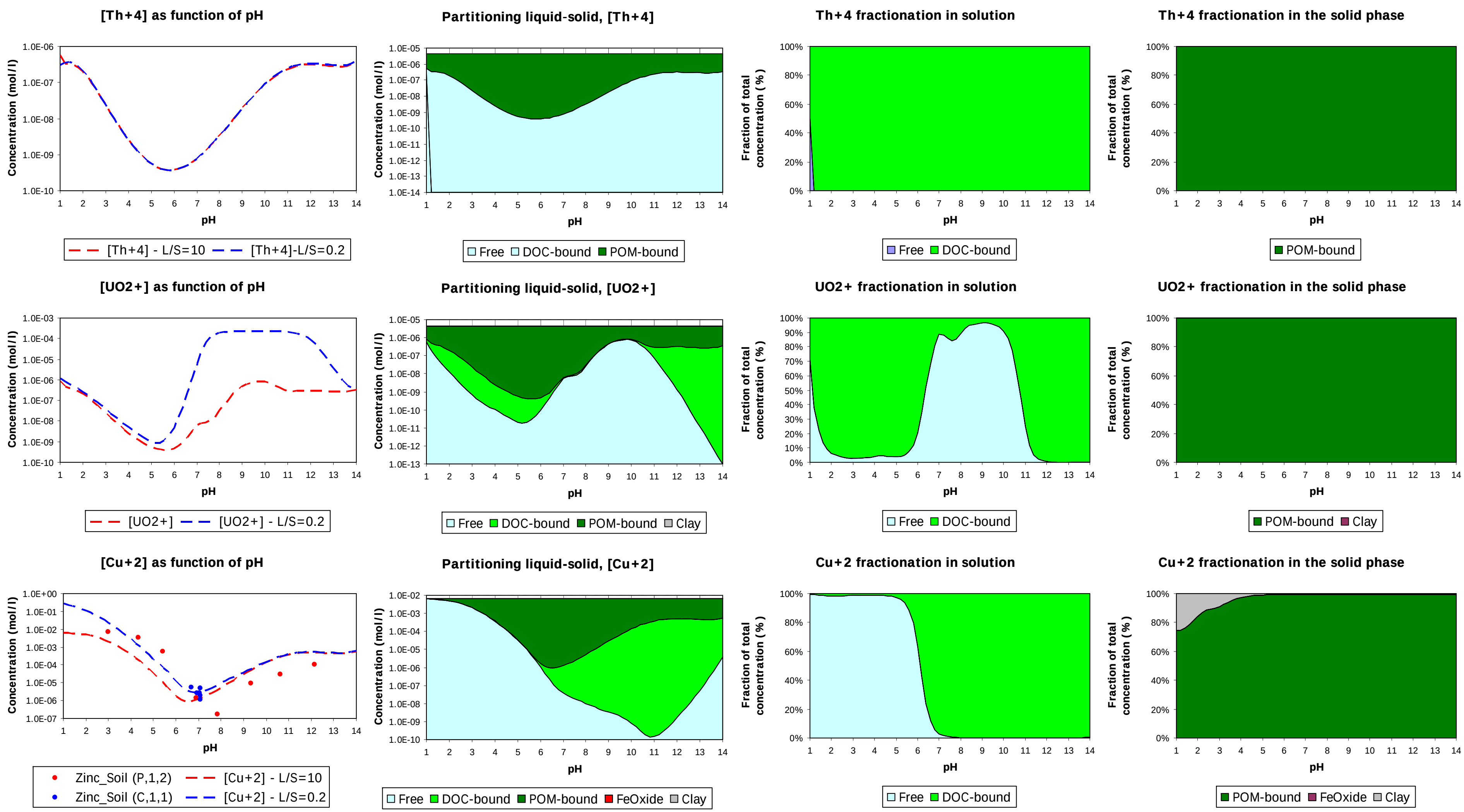
CLAYEY SOIL



Soil system dominated by dissolved and particulate organic matter interaction

PROFILE WASTE- CEMENTITIOUS BARRIER - SOIL

Input specification

Diffusion Case Stabilised Waste - CEM II GBFS-FA - Soil OXIDISED/ CARBONATED

Layer overview

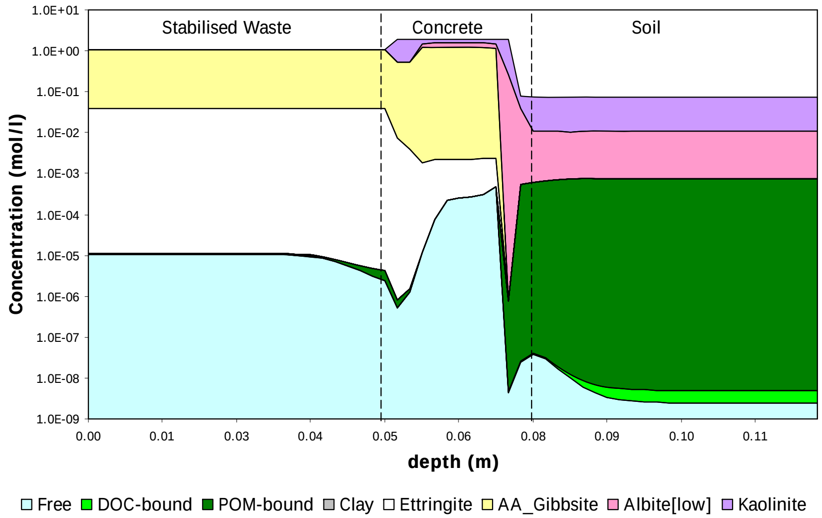
Material	Stabilised waste	GBFS-FA-Mortar	Soil	
Length	5.00	2.00	5.00	**cm**
Porosity frc	0.40	0.10	0.35	
Tortuosity	3.00	10.00	2.00	
Density	1.70	2.40	1.70	**kg/dm³**
pH	10.1	11.5	6.5	
pe	15	15	15	

In the modeling mineral dissolution, sorption on hydrated ironoxides, clay interaction, interaction with particulate and dissolved organic matter and incorporation in ettringite solid solution.

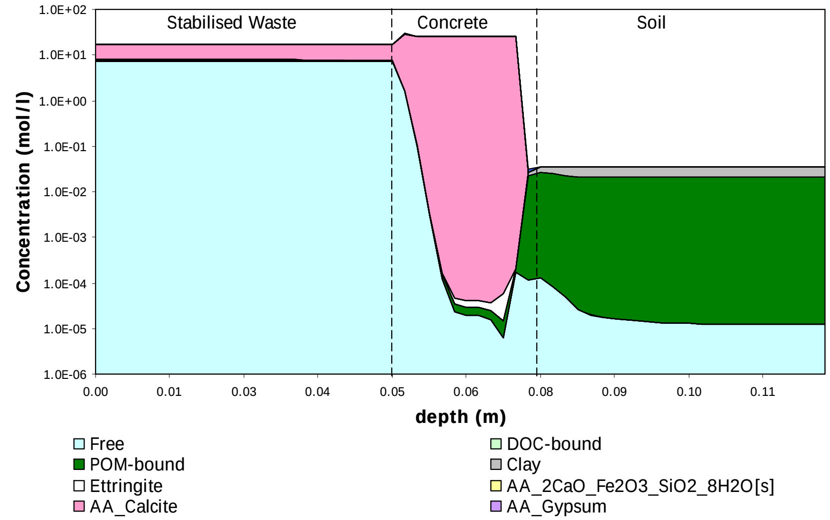
Typically 44100 variables, 192565 expressions, 118 equations

PROFILE WASTE- CEMENTITIOUS BARRIER - SOIL

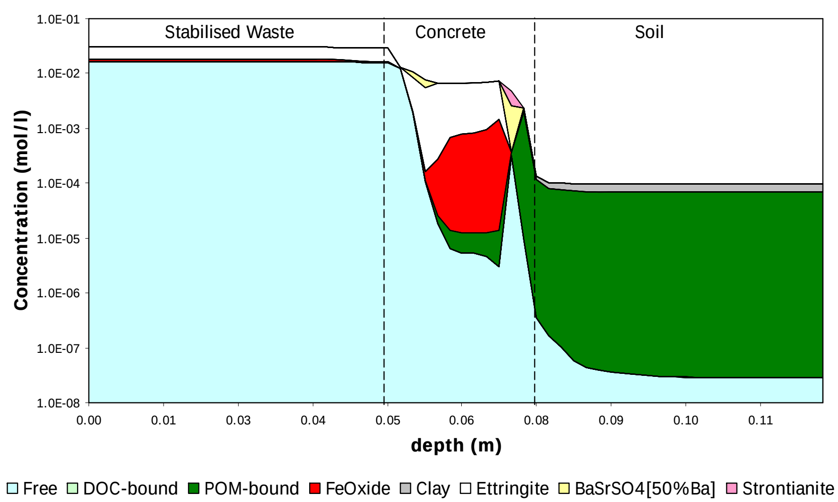
Distribution profile for Al+3 after 6 days



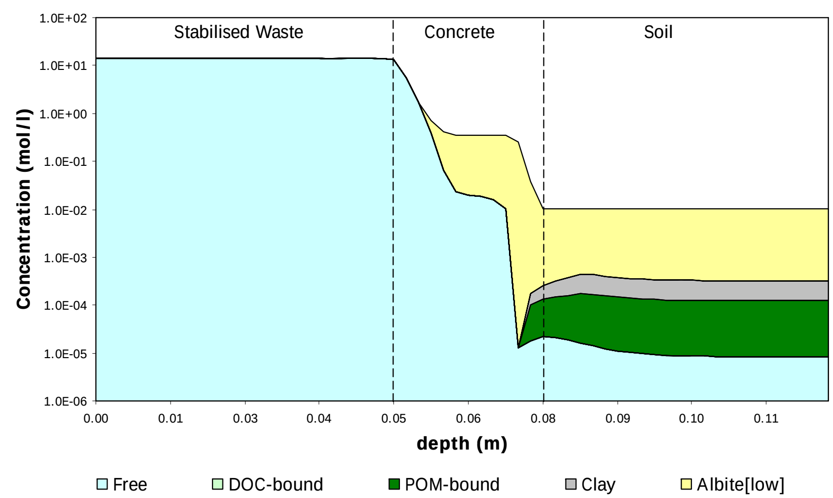
Distribution profile for Ca+2 after 6 days



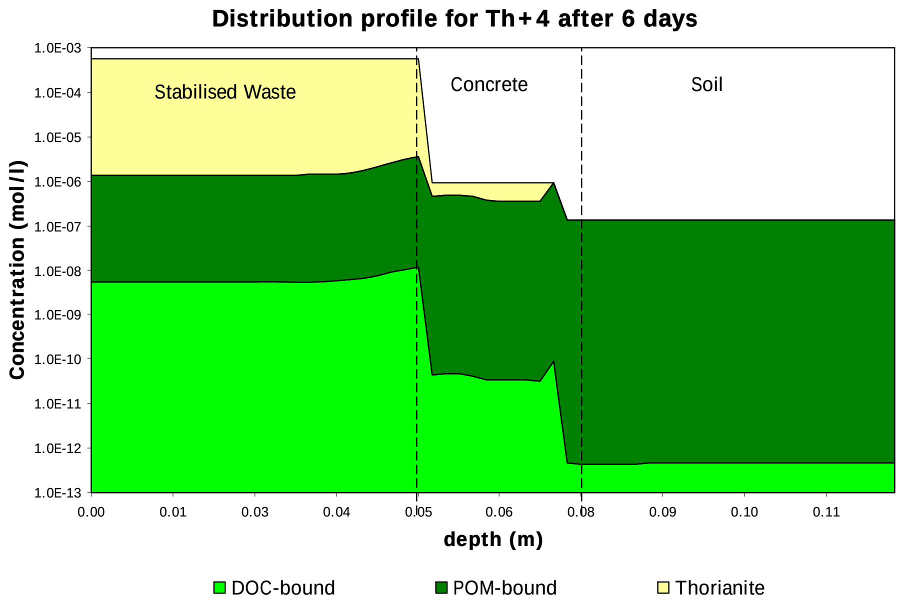
Distribution profile for Sr+2 after 6 days

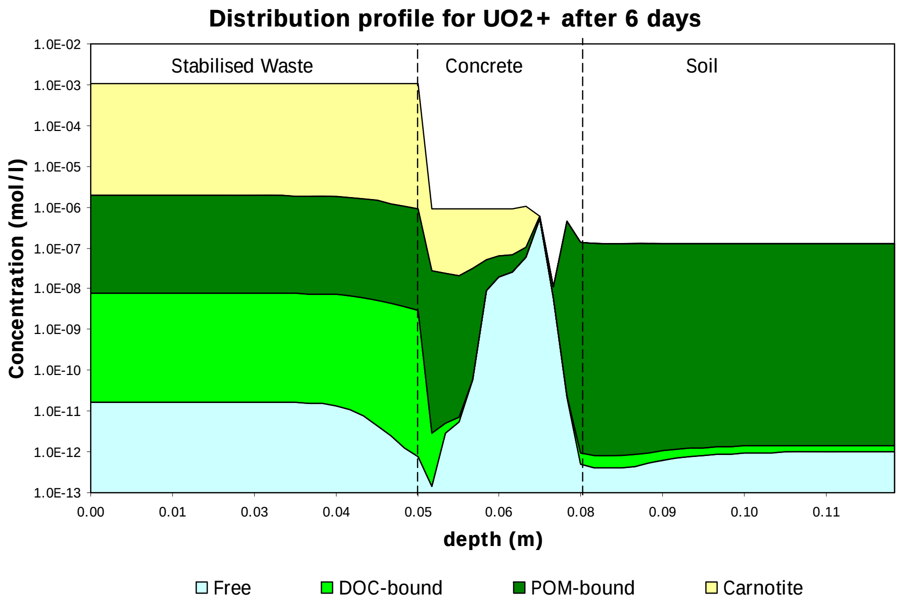


Distribution profile for Na+ after 6 days



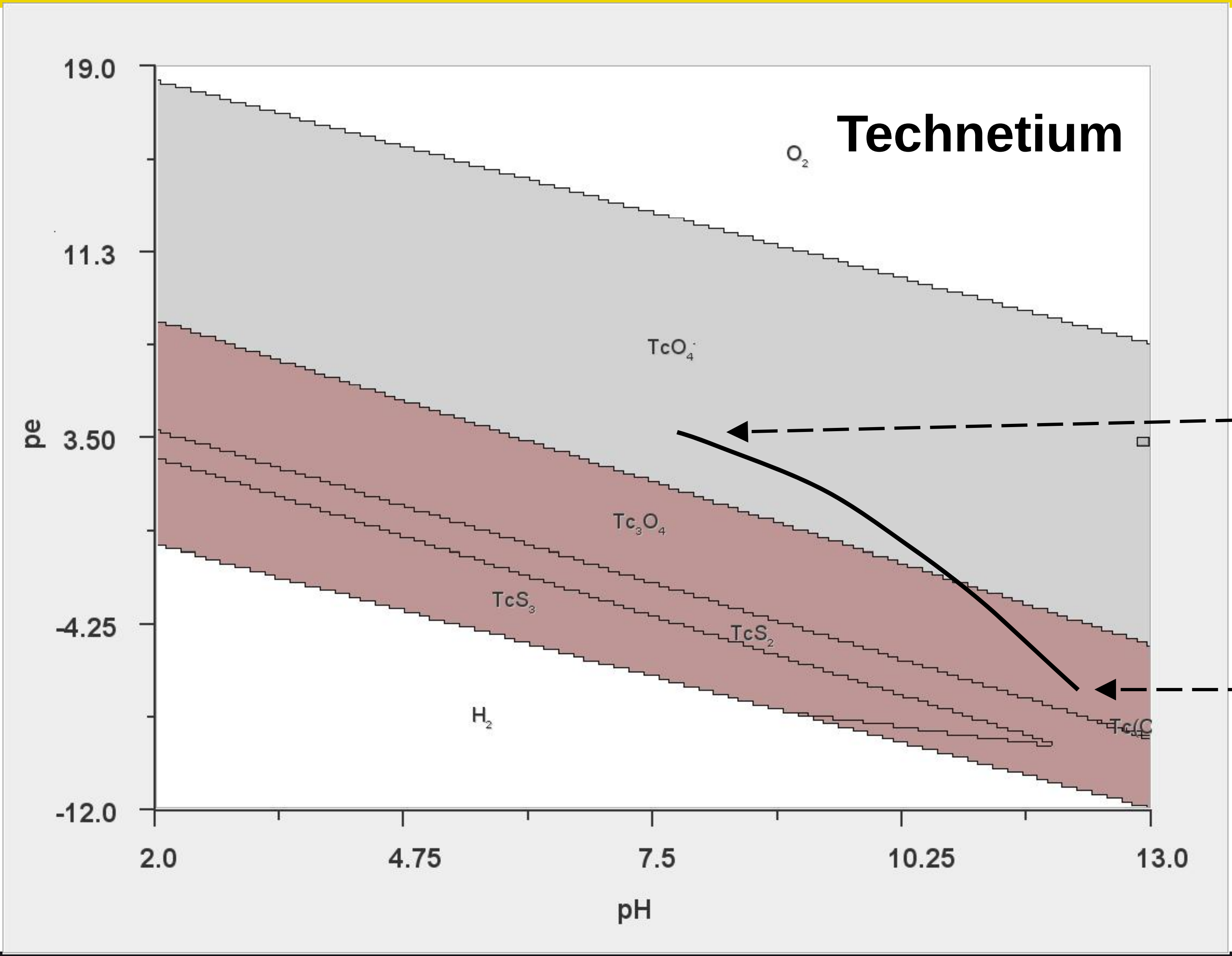
PROFILE WASTE- CEMENTITIOUS BARRIER - SOIL





Association with DOC important for release

Predominance diagrams

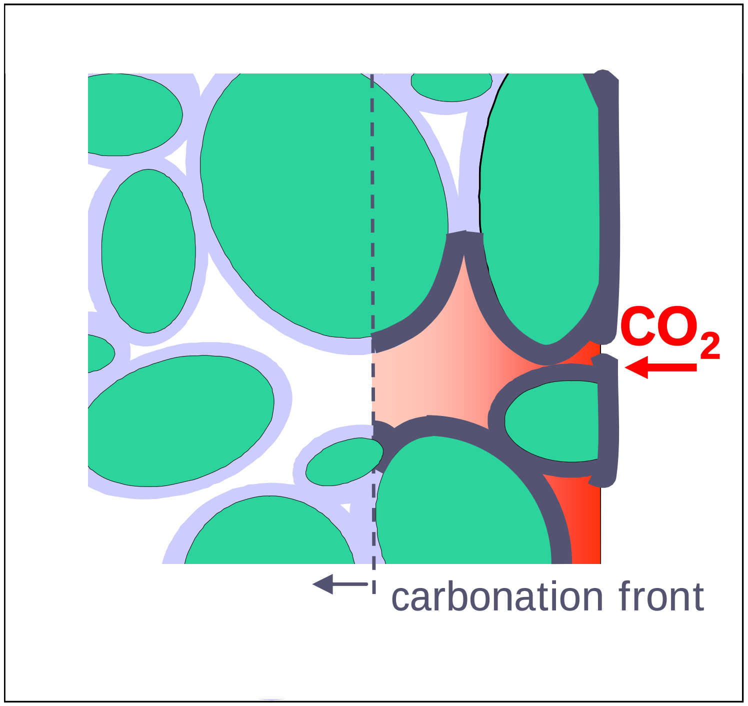


Projected very long term condition – carbonated and oxidised

Initial condition – alkaline and reducing

Carbonation leads to alterations in the release behaviour as a result of the pH change that is brought about

Carbonation



The effect of carbonation on release is illustrated by modeling, including an evaluation of uncertainty in the model prediction.

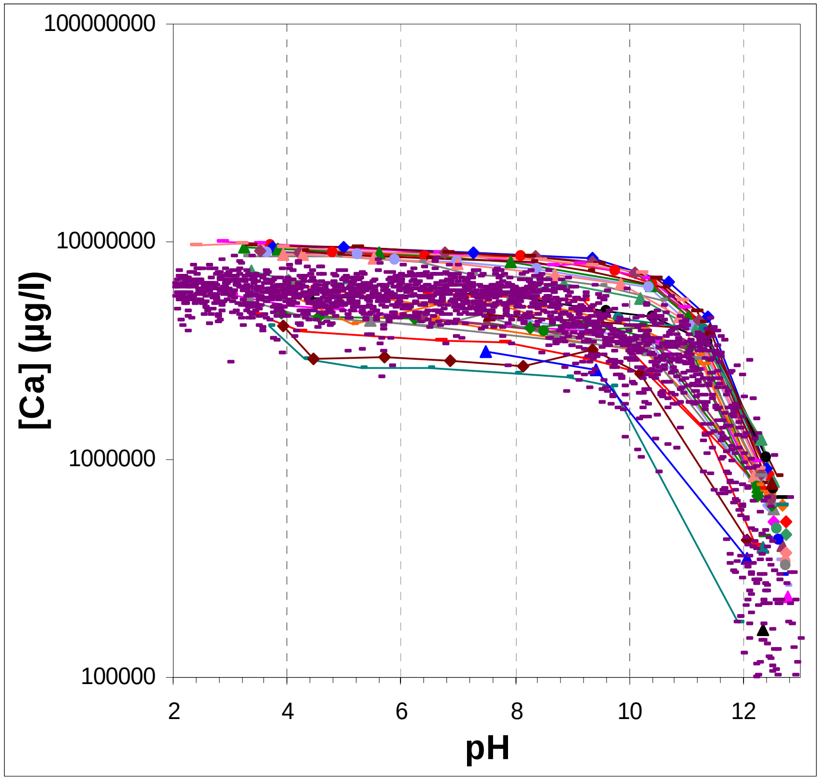
The data are placed in perspective to actually measured test data for > 70 different cement mortars (Portland as well as different types of blended cements)

Stochastically varied input parameters for modeling of pH dependence leaching test data for cement mortars

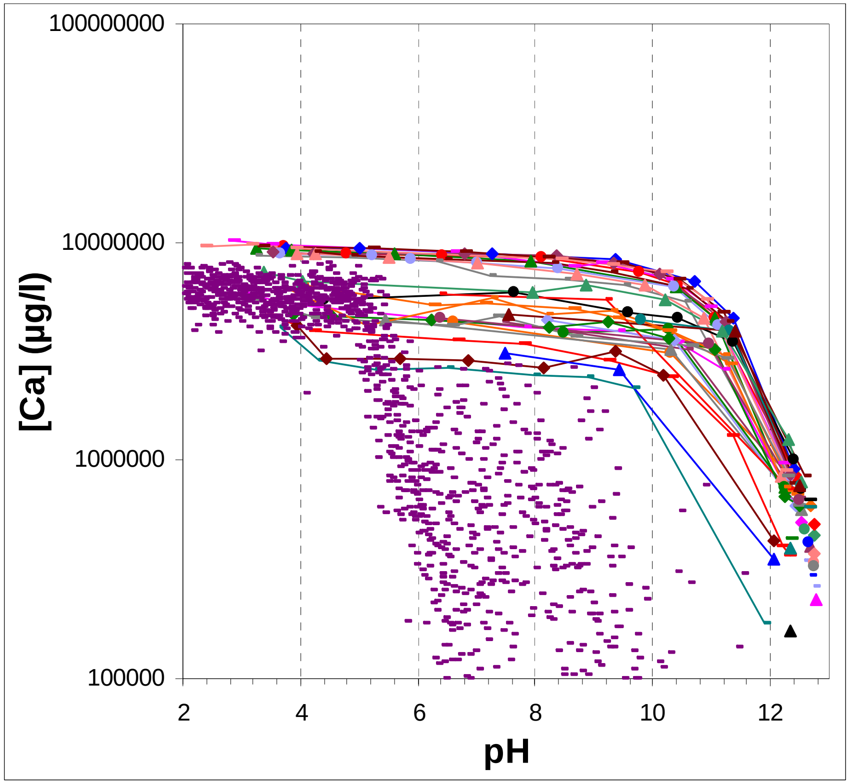
- Total available concentration (10%)
- pH (0.1 unit)
- Pe (2 units)
- All reaction constants (15%)
- Ionic strength (20%)
- Gaussian distribution
- 2000 simulations in the pH range 2-13

Solubility of Ca in cement mortars as function of pH

Without Carbonate

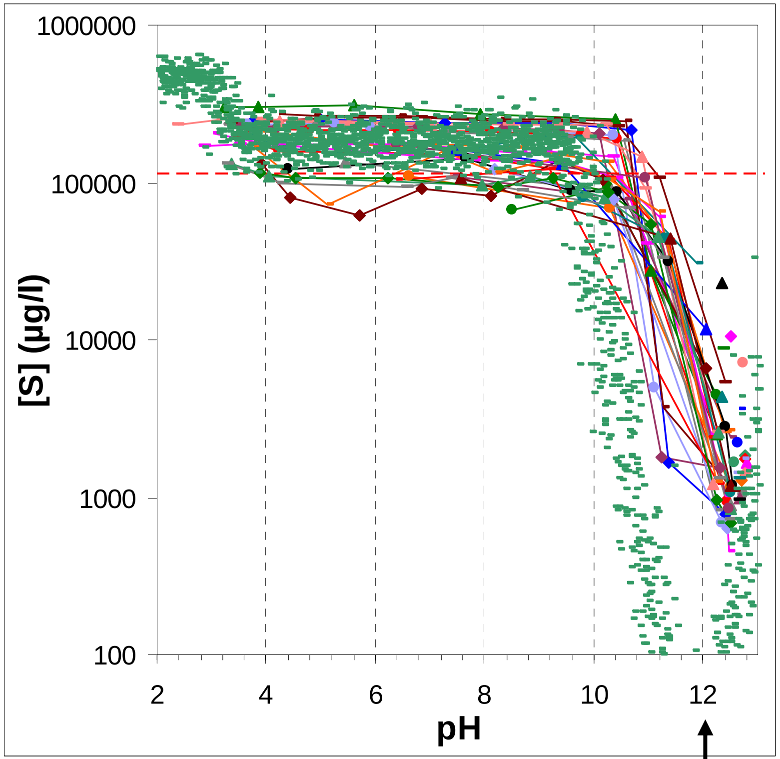


With Carbonate

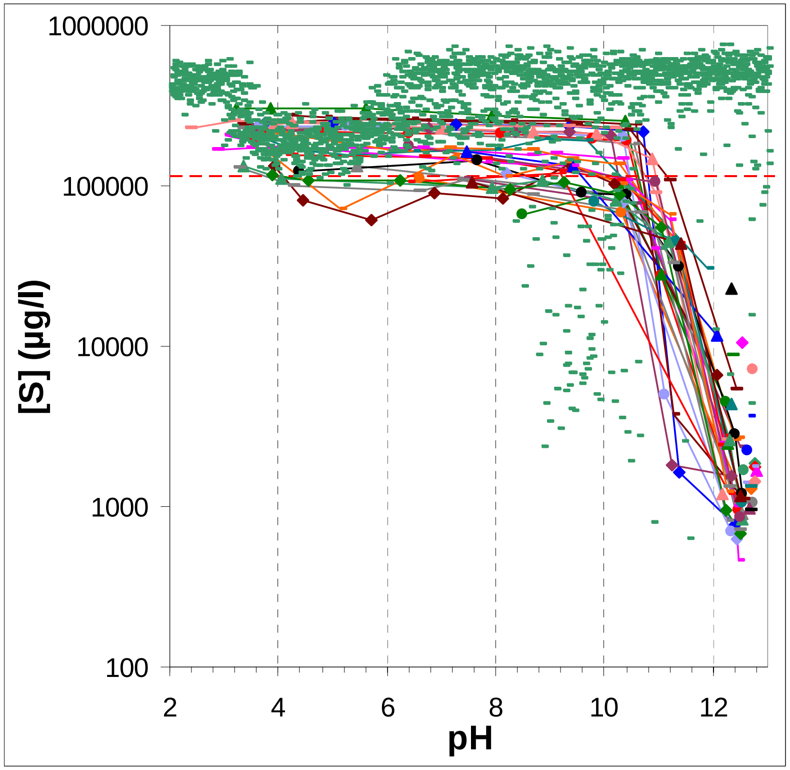


Solubility of SO4 as S in cement mortars as function of pH

Without Carbonate



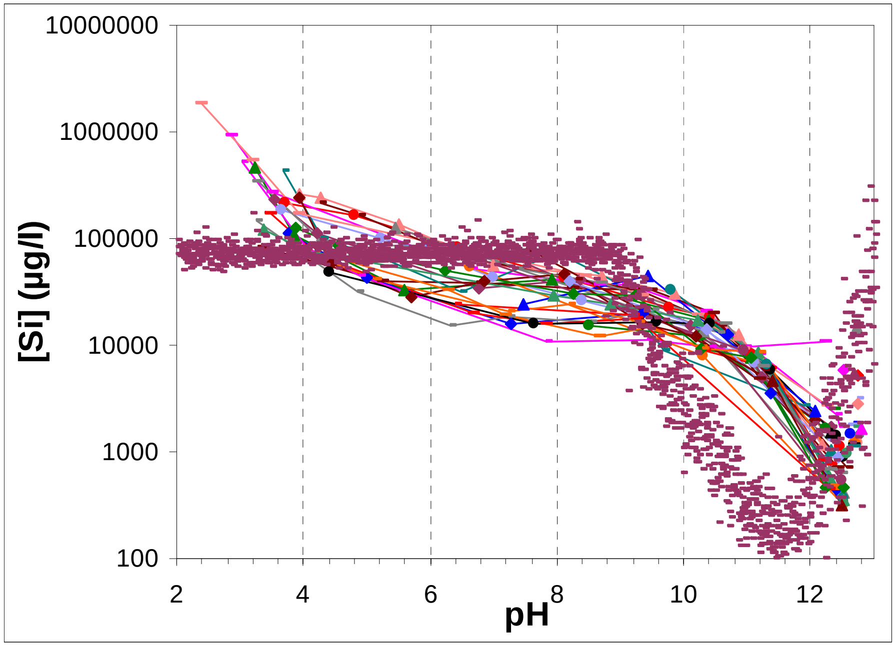
With Carbonate



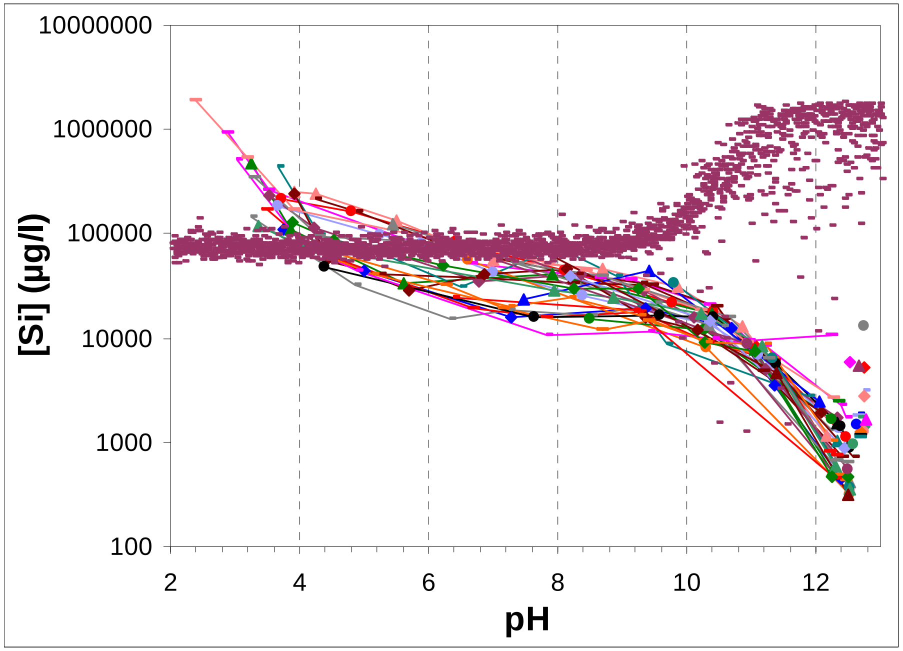
Ettringite

Solubility of Si in cement mortars as function of pH

Without Carbonate



With Carbonate



CONCLUSIONS

- Both physical and chemical changes in the waste form and cement barrier are of importance to properly assess release to the environmental.

- For non-reacting species a steady state condition of release through the barrier develops within a few years for saturated conditions. For unsaturated conditions this takes in the order of a hundred years.

- Carbonation and oxidation lead to important changes in release behaviour of substances. As these processes lead to moving fronts it is difficult to capture the release in a Kd describing contaminant behaviour of the entire waste form, the barrier or the soil.

CONCLUSIONS

- Gaining insight in more detailed chemical interactions is of importance as mobilisation in the form of dissolved complexes may occur. Currently, organic matter interaction is not considered.

- The binding potential of hydrated ironoxide (formed in situ upon oxidation of reduced Fe in both waste and barrier) for radionuclides of interest is important for retention within the containment under oxidised/carbonated conditions.

- U and Th in the present model runs are preliminary and need further verification by measurement of actual release behaviour from size reduced stabilised waste. In case of U and Th, this is possible with stable isotopes. For Tc this is obviously not possible.

- Therefore, carrying out a pH dependence test on cement stabilised radioactive waste is highly recommended to provide better insight in release controlling processes.

CONCLUSIONS

- More detailed chemical characterization provides the means to design for retention of contaminants in the waste or the design of a chemical barrier in addition to physical containment.

- Although calculation times with complex chemistry are long compared to Kd type calculations, it is possible to model release under defined conditions along the projected path as defined in a pe – pH diagram (resulting from carbonation and oxidation). More complete consideration of chemical processes also provides more robust understanding of non-linear process coupling and for improved design.

- Optimization of calculation efficiency. Balance complex models with simplified models. Preferably justified simplification based on understanding the underlying processes.