Probabilistic modelling of radionuclide release from a cementitious near field

T.G. Heath, B.T. Swift, S.A. Myers and A.R. Hoch

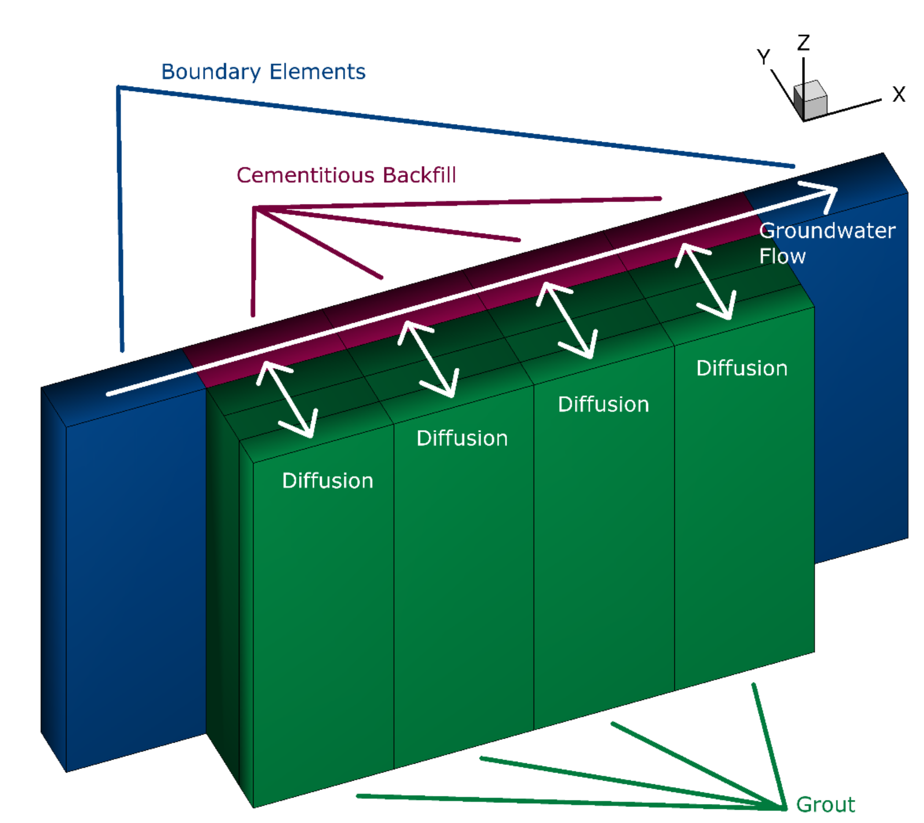
Wood, Building 150, Harwell Oxford, Didcot, Oxfordshire, OX11 0QB, UK.

Abstract

The IGD-TP's Strategic Research Agenda states that the safety case should describe the evolution of the repository as a reasonable representation of what might happen, and it should give a clear indication of uncertainties in the description. In the UK, this uncertainty requirement has led Radioactive Waste Management (RWM) to implement a probabilistic approach in the **Total System Model (TSM)** that underpins its safety case [1]. This approach involves assessing the uncertainty distributions for relevant input parameters in the TSM and then applying a Monte Carlo approach. This allows the identified input uncertainties to be propagated through to uncertainty in the calculated risk. However, where time-dependent parameters are approximated with constant values, real time variability is inevitably wrapped into the assessed uncertainty distributions. This approximation can result in the sampling and application of parameter values that are not appropriate to some ranges of evolved conditions, potentially leading to inaccurate results. To understand the effects of this assumption, the model needs to be refined to represent key processes in terms of time-dependent parameters [2].

This issue is particularly significant for cementitious intermediate-level waste concepts in higher-strength rocks, where near-field solubility and sorption parameters for radionuclides may vary owing to evolving chemical conditions. In the TSM, these parameters have been treated as time-independent quantities. To improve on this assumption, a **Near-field Component Model (NFCM)** has been developed that provides a more detailed and realistic representation of the system within a framework of the probabilistic Monte Carlo approach. The groundwater chemistry, radionuclide chemistry and backfill reactions are represented explicitly as chemical equilibria. The associated radionuclide equilibrium constants are treated as probabilistic parameters in the NFCM, with uncertainty distributions defined by their previously assessed standard deviations. Comparing the outputs of the NFCM and TSM shows that the results are very similar where the radionuclide chemistry remains constant, e.g. for uranium(IV), but differ significantly where the chemistry varies, e.g. for uranium(VI).

Numerical Modelling



The **NFCM** combines:

- the probabilistic capabilities of the Monte-Carlo simulation program GoldSim, with
- the detailed chemistry and flow capabilities of the reactive transport program TOUGHREACT.

Schematic of the mesh with 18 grid blocks, representing groundwater flow through backfill and diffusion into adjacent grout

Radionuclide Chemistry

The **NFCM** represents the chemistry of the radionuclides, groundwater and cementitious solids as equilibrium reactions. The chemistry of the major elements is treated deterministically. The radionuclides are characterised by:

- Probability Density Functions (PDFs) created from $\log_{10}K°$ and uncertainty values (from ThermoChimie, NEA);
- PDFs for solubility-limiting phases derived using the same assumptions as those underlying the TSM data elicitations [3].

But care is needed to identify any parameter correlations and to avoid unnecessary error inflation.

Physical parameters

Probabilistic: groundwater flow rate.

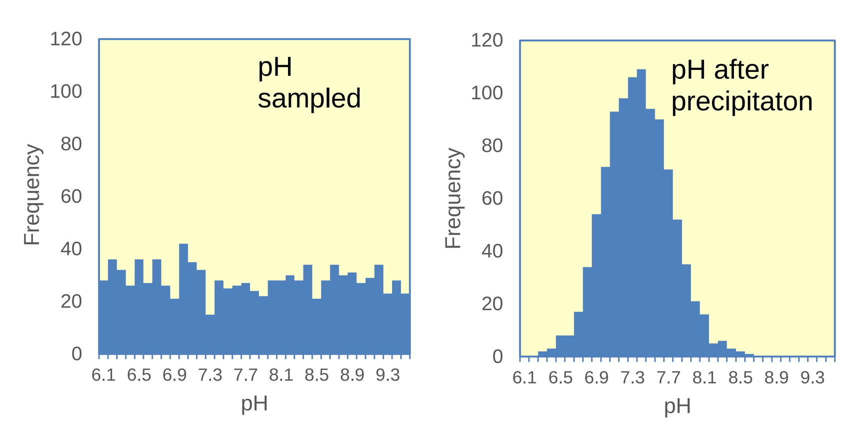
Deterministic: diffusion coefficient, tortuosity, permeability, porosity, area between grout and backfill.

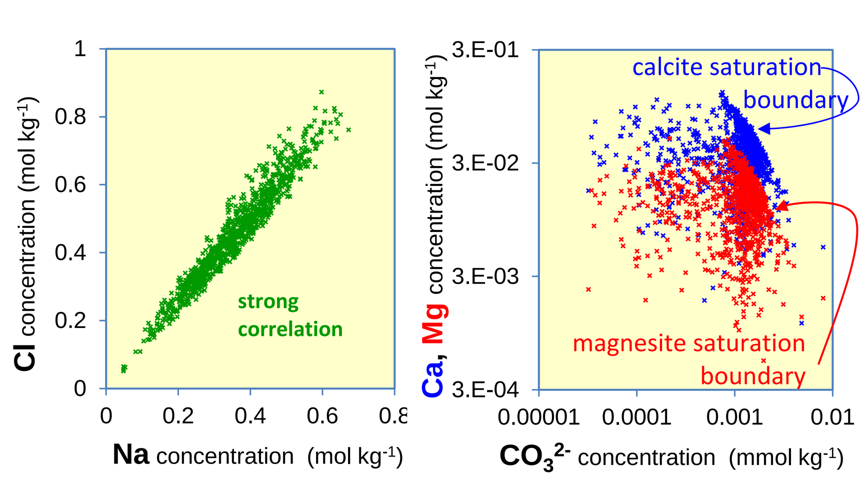
Porosity evolves and affects transport.

Groundwater Chemistry

The **NFCM** is illustrated for a generic hard-rock geology, so the associated groundwater uncertainty is large:

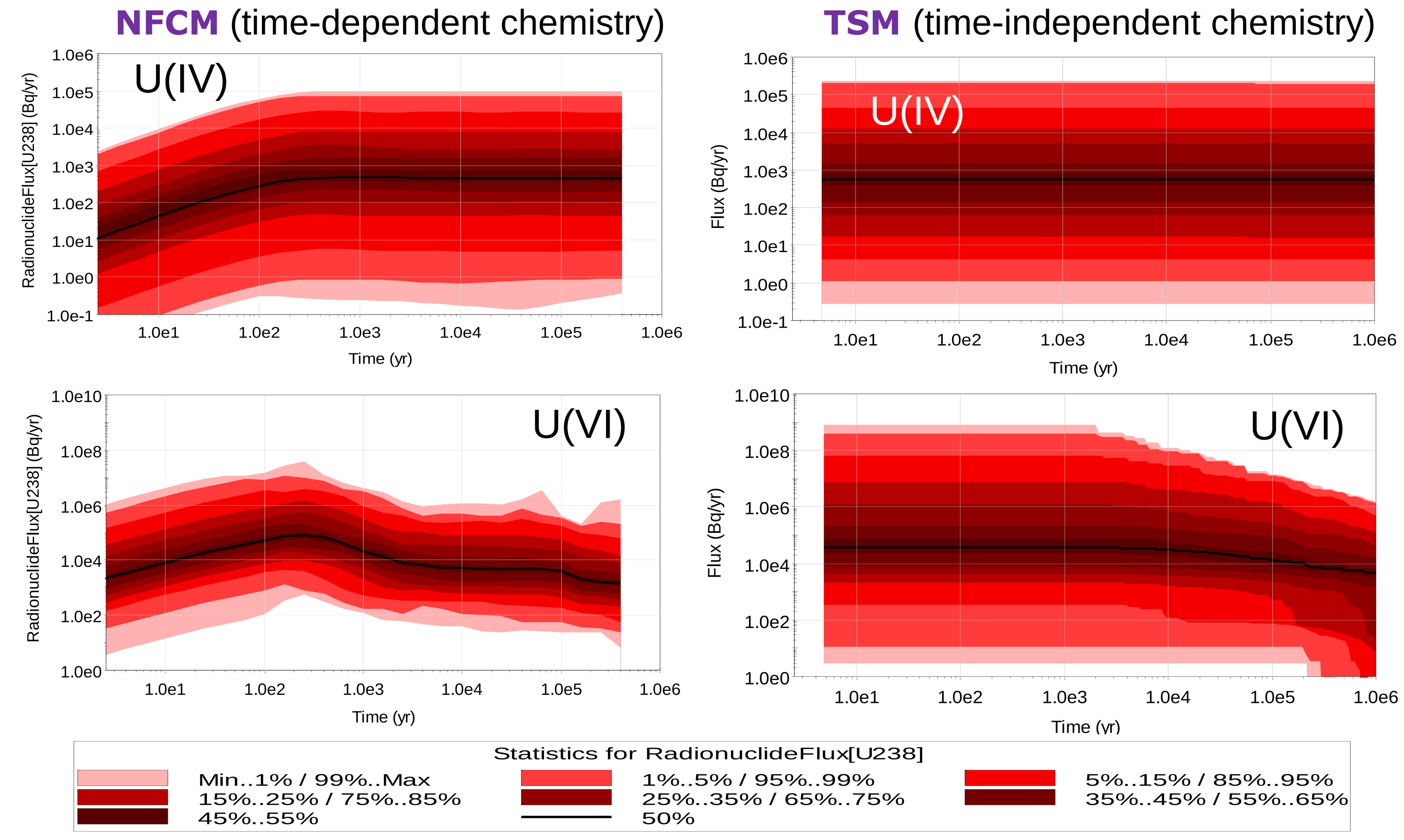
- sampled groundwaters are generated by random mixing of four end-member compositions;
- precipitation of simple solids is included.





Model Results

NFCM (time-dependent chemistry) **TSM** (time-independent chemistry)

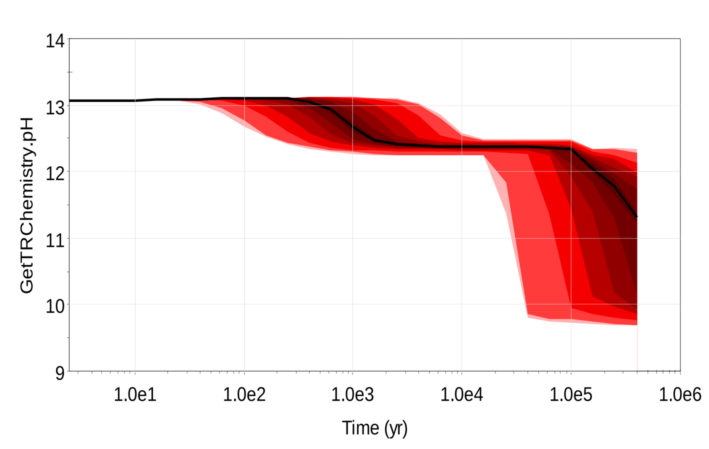


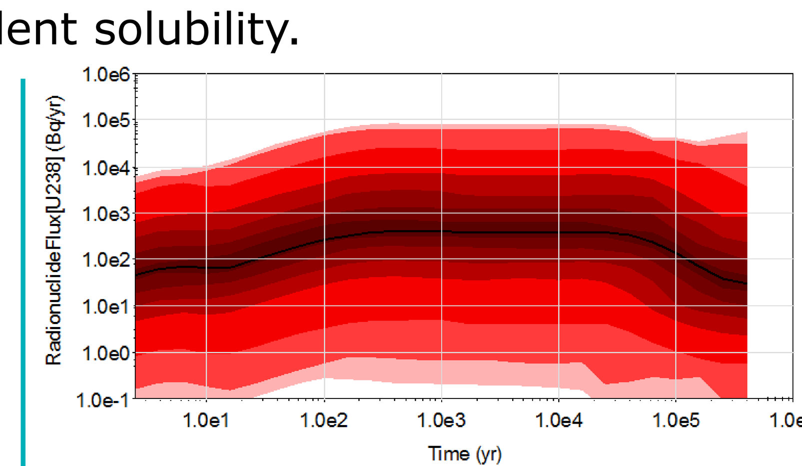
Radionuclide fluxes from near field (contour plots - 200 realisations)

Results are shown for uranium fluxes under solubility control, for U(IV) under strongly reducing conditions and U(VI) under mildly reducing conditions.

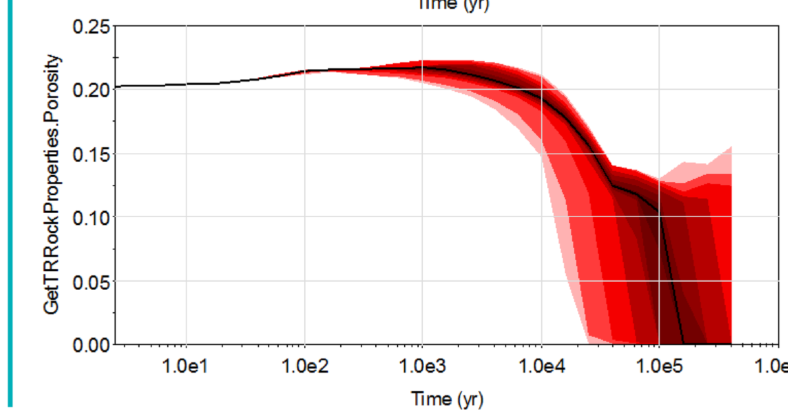
- The **NFCM** results show an early transient with flux increasing as uranium diffuses from the grout into the backfill where groundwater is flowing (whereas the **TSM** one-box model allows immediate flux).
- At longer times, U(IV) shows similar behaviour since the key equilibrium is not sensitive to evolving conditions: $UO_2.2H_2O(am) = U(OH)_4(aq)$.
- In the **NFCM**, U(VI) shows an evolving flux owing to its complex chemistry and time-dependent solubility.

Spread of pH in the downstream backfill grid block (NFCM)





Effect of allowing for porosity changes on U(IV) flux



Spread of porosities in downstream grout adjacent to backfill

Conclusions

The **NFCM** has been developed to extend the probabilistic approach used in UK performance assessments to the explicit treatment of uncertainty in radionuclide and groundwater chemistry. Compared with the TSM, which uses time-independent radionuclide solubilities, it has shown:

- **For radionuclides with constant speciation** under evolving conditions, e.g. U(IV), Pu(IV), mean radionuclide fluxes and their spreads agree well between results from the **NFCM** and the **TSM**;
- **For radionuclides with varying speciation**, e.g. U(VI), significant differences are found:
  - fluxes are time dependent and vary with evolving conditions in the grout and backfill
  - the spread of fluxes from the near field is typically narrower than that for the TSM
  - lower solubilities in the grout reduce the release from the near field below that expected for backfill solubility-controlled limits (as assumed in the TSM).

In the future, inclusion of the effects of cellulose degradation products (especially isosaccharinic acid) is expected to result in larger differences.

References

1. Nuclear Decommissioning Authority, Geological Disposal: Generic Post-closure Safety Assessment, NDA Report NDA/RWMD/030, 2010.
2. RWM, Geological Disposal: Methods for Management and Quantification of Uncertainty, NDA/RWM/153, 2017.
3. D. Swan and C.P. Jackson, Formal Structured Data Elicitation of Uranium Solubility in the Near Field, Serco Assurance Report, SA/ENV/0920 Issue 3, 2007.

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Contact

Tim Heath: tim.heath@woodplc.com