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Case Study

Modelling radionuclide transport in fractured media with a dynamic update of K_d values

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ABSTRACT

Radionuclide transport in fractured crystalline rocks is a process of interest in evaluating long term safety of potential disposal systems for radioactive wastes. Given their numerical efficiency and the absence of numerical dispersion, Lagrangian methods (e.g. particle tracking algorithms) are appealing approaches that are often used in safety assessment (SA) analyses. In these approaches, many complex geochemical retention processes are typically lumped into a single parameter: the distribution coefficient (K_d). Usually, the distribution coefficient is assumed to be constant over the time frame of interest. However, this assumption could be critical under long-term geochemical changes as it is demonstrated that the distribution coefficient depends on the background chemical conditions (e.g. pH, Eh, and major chemistry). In this work, we provide a computational framework that combines the efficiency of Lagrangian methods with a sound and explicit description of the geochemical changes of the site and their influence on the radionuclide retention properties.

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1. Introduction

Crystalline rock formations are under consideration by several countries as host formations for geological disposal of radioactive wastes. Over the long time frames of interest in assessing safety of those potential disposal systems, environmental conditions are expected to change. Such changes are expected to be particularly dramatic for repositories located at high latitudes. For example, the prospective Finnish and Swedish repositories (Posiva, 2013; SKB, 2013) for spent nuclear fuel are expected to see climatic changes from temperate to periglacial and eventually to glacial conditions over a 100,000 year period. Such changes will significantly alter the hydrological and geochemical conditions in the host rock.

A key component of the aforementioned safety assessment (SA) studies is the analysis of possible releases of radionuclides from a canister in a deposition hole and their transport along preferential flow paths in the fractured rock. This analysis is typically carried out by means of radionuclide transport simulations. In the last Swedish and Finnish safety analyses (i.e. SR-Site, SKB, 2011, and TURVA2012, Posiva, 2012), most of these calculations were carried out using the time domain particle tracking code Migration Analysis of Radionuclides in the Far Field (MARFA) (Painter et al., 2008; Painter and Mancillas, 2013). MARFA uses a numerically efficient algorithm, which allows complex calculations to be carried out relatively quickly. This efficiency is particularly appealing in SA studies, where typically a large number of variant simulations are required. Besides computational efficiency, MARFA is free of numerical dispersion, which means that computed breakthrough curves are not affected by spurious effects such as artificial dilution. A summary of MARFA's processes and conceptualizations and of the variant versions developed for this work is presented in Appendix A.

As in most of the Lagrangian-based transport codes (e.g. Salamon et al., 2006, and references there in), in MARFA sorption reactions are represented using distribution coefficients (K_d), which are usually kept unchanged over the simulation time frame. It is worthwhile noting that K_d-based transport calculations are not suited to describe chemical retention processes other than sorption (e.g. the precipitation of radionuclides in minerals or solid solutions).

The dependence of specific contaminant distribution coefficients on the underlying background geochemical conditions (i.e. pH, redox conditions, salinity, and TDS) has been widely studied

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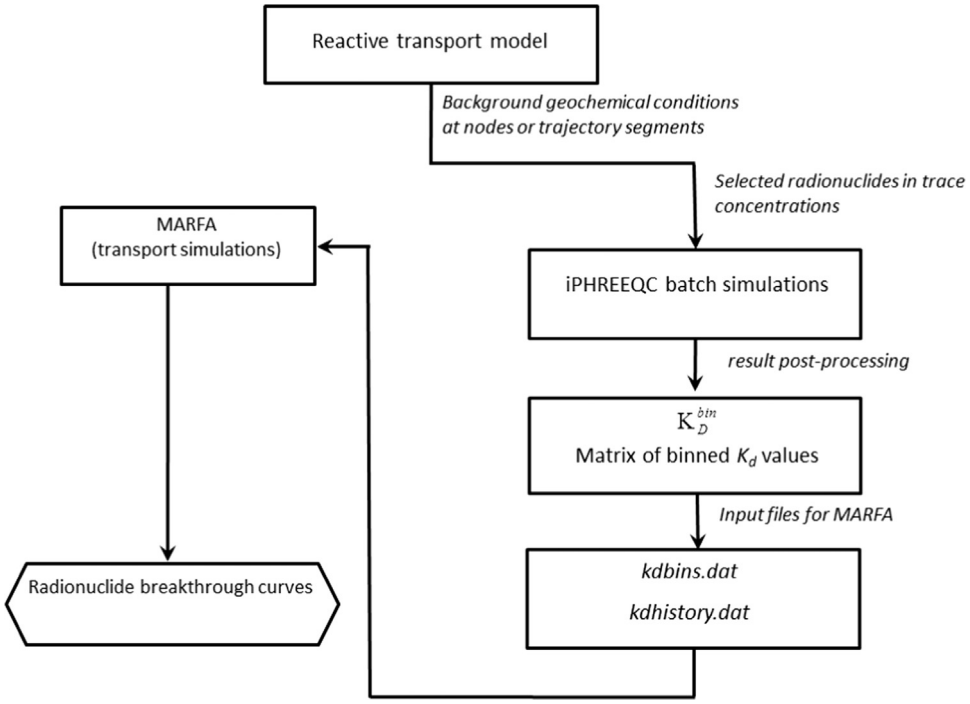

Fig. 1. Functionality overview of iFM.

and documented (e.g. Abril, 1996; Sauve et al., 2000; Crawford et al., 2006; Granizo and Missana, 2006; Domènech et al., 2015). Different numerical studies have made attempts to keep track of this dependency in K_d-based radionuclide transport simulations using the so-called "smart K_d" approach (e.g. Stockmann et al., 2012; Flügge et al., 2013; Noseck et al., 2014). In these works the computationally expensive reactive transport calculations that are needed to determine spatial and temporal variations in K_d's are performed using simplified geochemical calculations, where environmental markers (e.g. H^+ and ionic strength) are transported and affected by simplified reactions. These calculations are coupled to the K_d-based radionuclide transport simulations.

Here we propose an alternative and novel formulation of the "smart K_d" approach, which relies on two basic premises: (i) the evolution of the background geochemistry need to be described in its full complexity as "individual sub-processes cannot be fully assessed without considering them in the context of the other dynamic processes" (Steefel et al., 2005) and (ii) radionuclide transport calculations need to be efficient and free of numerical dispersion.

In the proposed methodology, the mutual interplay between physical and (bio)geochemical processes and their effect on the hydrochemical conditions of the site under study are captured by means of mechanistic multicomponent reactive transport calculations. This, which is an important difference of the methodology compared to the previously cited "smart K_d" approaches, allows the full complexity and non-linearity of the system to be consistently included in the numerical framework.

The requirements of numerical efficiency and absence of numerical dispersion are achieved by decoupling the simulation of radionuclide transport from the calculation of the background geochemistry, under the hypothesis that radionuclides are in trace concentration. This allows radionuclide transport calculations to be efficiently carried out using MARFA and this, in turn, allows specific uncertainties (e.g. release location and retention model) to be treated in a probabilistic fashion. Other types of uncertainties, related to the evolution of the background geochemistry, are more difficult to represent as the underlying model is built upon computationally expensive mechanistic reactive transport calculations.

2. Methodological approach

The basic premise of the approach is that the radionuclides are in trace concentration, which implies that (i) they do not affect the evolution of the background geochemistry and (ii) their retention is linear with concentration. Given this assumption, the methodology follows these steps:

1. The evolution of the background geochemistry is computed using a mechanistic flow and reactive transport code.
2. The computed chemical solutions (i.e. concentration of aqueous species, including redox conditions and temperature, at each node of the domain and at each considered time step), the selected sorption model and an arbitrarily low radionuclide concentration are used to carry out mechanistic 0D batch calculations.
3. The results provide a database of distribution coefficients (in space and time) that is used to dynamically update the K_d values used in the MARFA radionuclide transport simulation.

The retention model (Step 2) is defined by the user and has to be representative of the actual mineralogy of the site under study.

Steps 2 and 3 are run automatically using a numerical tool, denoted as iFM (interface to MARFA). iFM is a python script that takes care of the following operations:

A: The chemical solutions are retrieved from the results of step 1 and stored in memory.
B: The sorption model is read from the user configuration file.
C: For each chemical solution, a batch calculation is performed using the dynamic library IPhreeqc (Charlton and Parkhurst, 2011). In the calculation, an arbitrarily low radionuclide concentration is added to the solution and the resulting aqueous composition is equilibrated with the sorption sites.

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