An Equivalent Kd-based Radionuclide Transport Model Implemented in COMSOL Multiphysics® Software

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Abstract

The Swedish Nuclear Fuel and Waste Management Company (SKB) has developed conceptual and quantitative models for the reactive transport of selected radionuclides in the near-surface system or regolith (Grandía et al., 2007; Sena et al., 2008; Piqué et al., 2010). However, the description of hydrological processes and the model geometries were simplified, due to computational constrains and the limitations of the available reactive transport tools

Radionuclide sorption is often simulated using a lumped approach where retention processes are represented by a single parameter, the distribution coefficient (Kd), which relates the radionuclide mass retained in the solid phase to its aqueous concentration. Kd-based simulations rely on two strong assumptions: Kd depends on soil properties and is constant in time. However, sorption processes depend also on the fluid characteristics, which can be spatially and temporally variable.

Here, a 3D mechanistic reactive transport model of the regolith was implemented in iCP (Nardi et al., 2014). The result of this model provides a dataset of Kd values representative of the actual geochemical conditions of the regolith. That dataset is used to build equivalent 3D Kd-based models describing the retention of 90Sr, 137Cs, 235U and Ra. The model was implemented in Comsol using the Darcy's Law (Subsurface Flow module, Fluid Flow), and Solute Transport (Chemical Species Transport module) physics and accounts for steady state saturated-unsaturated flow assuming constant recharge and radionuclide transport with linear sorption. Also, high resolution of 3D hydrological and geological conditions in the regolith is considered.

Two types of Kd interpretation were compared: "equivalent Kd spatial field" versus "2-single time dependent Kd values". The first approach determines Kd values using the concentration distributions of dissolved (Figure 2a) and retained (Figure 2b and c) radionuclides at the end of the iCP simulation. This generates a unique spatial field for each radionuclide (Figure 2d). The second approximation determines a spatially averaged Kd value for each layer that varies with time. This generates a temporal Kd function for the till material and another one for the clay (Figure 2e).

Overall, breakthrough curves show discrepancies between both equivalent Kd-based transport models and the fully reactive transport model (Ra and Sr present some better fit). The 137Cs evolution suggests that the 2-single time dependent Kd values approach is better than the equivalent Kd spatial field approach (Figure 3a and 3b). Spatial distributions of retained radionuclides and their temporal evolution show that the formulation based on 2-single time dependent Kd values reproduces the retention behavior of 90Sr and 137Cs (Figure 3c) better than the equivalent Kd spatial field approach, while this reproduces the retention behavior of 235U and Ra (Figure 4) better than the former. This is because the dominant retention processes of 90Sr and 137Cs are mainly dynamic, while the main mechanisms of 235U and Ra retention undergo local spatial variations that dominate over the temporal changes.

The equivalent Kd-based transport model could be improved by approaches accounting for a dynamic update of the Kd values or by other alternative non-local in time and space approximations.

Reference

Grandía, F., Sena, C., Arcos, D., Molinero, J., Duro, L., Bruno, J., 2007. Quantitative assessment of radionuclide retention in the near-surface system at Forsmark. Development of a reactive transport model using Forsmark 1.2 data. SKB R-07-64, Svensk Kärnbränslehantering AB.

Nardi, A., Idiart, A., Trinchero, P., de Vries, L. M. and Molinero, J., 2014. Interface Comsol-PHREEQC (iCP), an efficient numerical framework for the solution of coupled multiphysics and geochemistry, Computers & Geosciences, doi:10.1016/j.cageo.2014.04.011.

Piqué, À., Grandía, F., Sena, C., Arcos, D., Molinero. J., Duro. L., Bruno. J., 2010. Conceptual and numerical modelling of radionuclide transport in near-surface systems at Forsmark. SR-Site Biosphere. SKB-R-10-30, Svensk Kärnbränslehantering AB.

Sena, C., Grandía, F., Arcos, D., Molinero, J., Duro, L., 2008. Complementary modelling of radionuclide retention in the near-surface system at Forsmark: Development of a reactive transport model using Forsmark 1.2 data. SKB-R-08-107, Svensk Kärnbränslehantering AB.

Figures used in the abstract

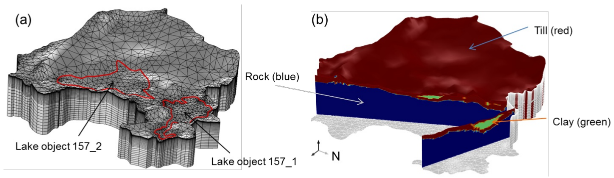

Figure 1: (a) 3D mesh indicating the location of the two discharge objects and the vertical discretization in the two blocks (vertical exaggeration x10z). (b) Geochemical model subdomains. The granitic rock (blue) is considered inert and the regolith is divided into till and clay domains.

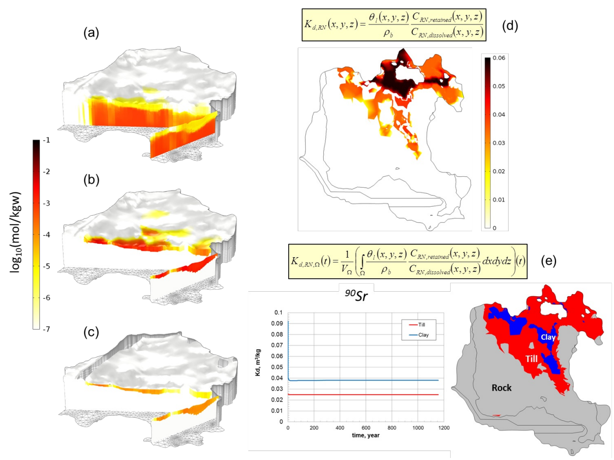

Figure 2: Kd interpretation methodology (retention of 90Sr): 3D Distributions of (a) dissolved 90Sr, (b) 90Sr adsorbed on planar sites and (c) 90Sr precipitated as strontianite are used to calculate the (d) equivalent Kd spatial field and the (e) 2-single time dependent Kd values.

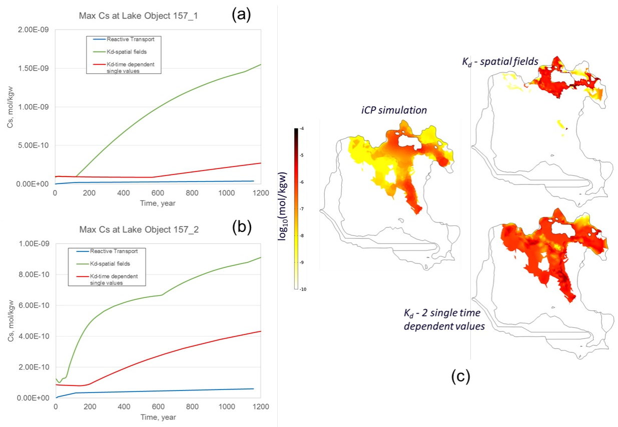

Figure 3: Evolution of the maximum concentration of 137Cs at the 157_1 (a) and 157_2 (b) lake objects. (c) Concentration of retained 137Cs simulated with iCP (fully reactive transport model) compared with the concentrations of retained 137Cs simulated with the equivalent Kd-based transport models (z = 3 m, after 1100 years of simulation).

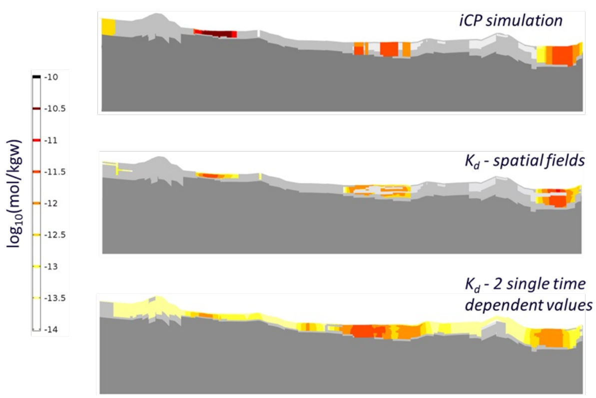

Figure 4: Concentration of retained Ra simulated with iCP (fully reactive transport model) compared with the concentrations of retained Ra simulated with the equivalent Kd-based transport models (Valley cross section, after 1100 years of simulation).