COMSOL CONFERENCE 2015 GRENOBLE

AMPHOS 21

SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

An equivalent K_d-based radionuclide transport model implemented in Comsol

Orlando Silva, Elena Abarca, Jorge Molinero, Ulrik Kautsky

October 15th 2015, Grenoble

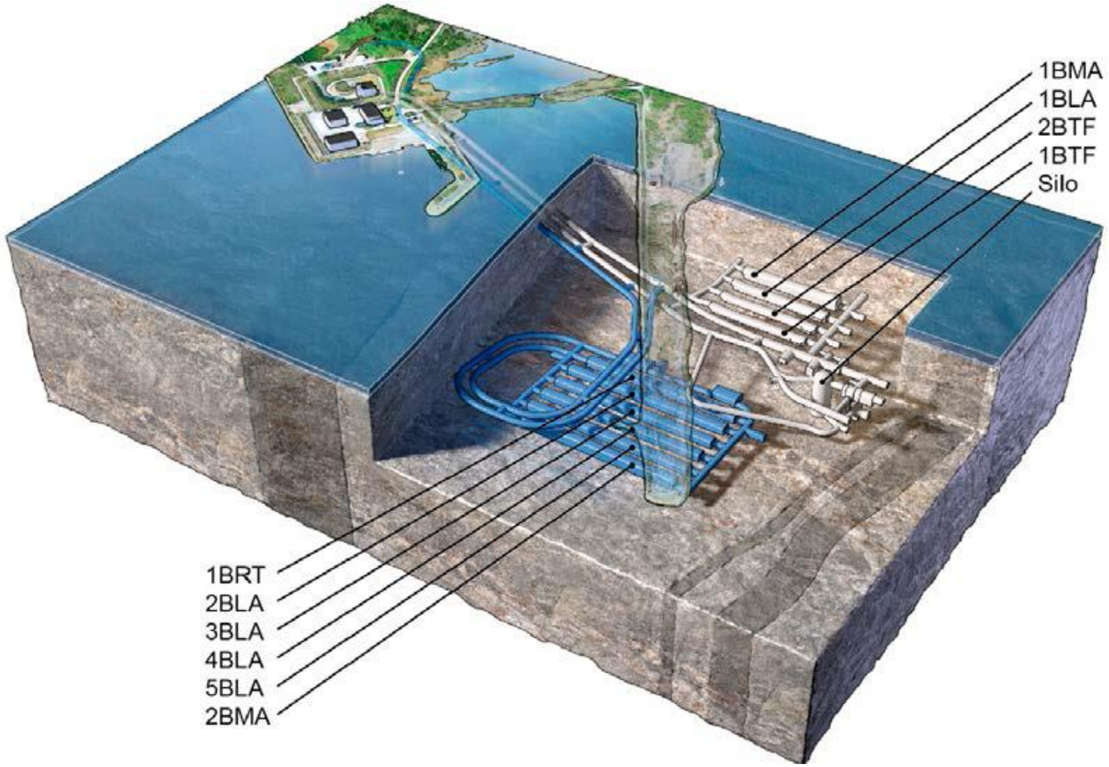
1994 - 2014 20 YEARS FLOWING

Outline

- Background
- Problem
- Objectives
- Modeling Approach
  - Fully reactive transport model – iCP
  - Retention processes
  - Linear sorption model – Comsol
  - Geometry, mesh and BC
  - Equivalent K_d approaches
- Results
- Conclusions

Background

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. Hydrological processes and geometries are complex → need of practical and powerful reactive transport tools.



SFR repository at Forsmark

Conceptual illustration of the hydrological system for different climate conditions (not to scale)

Problem

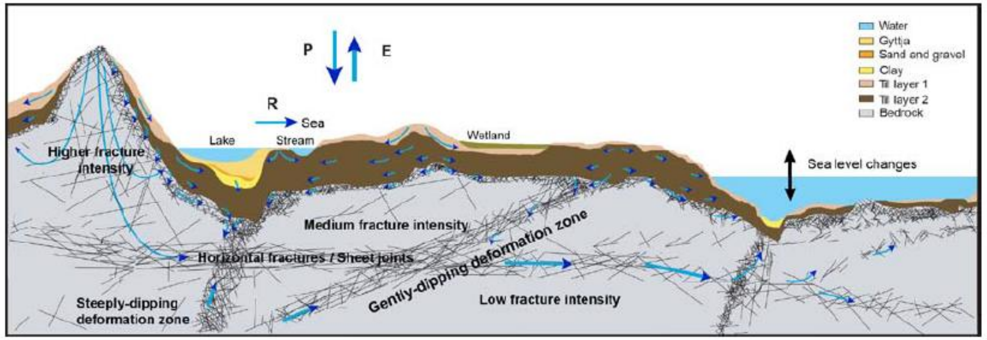
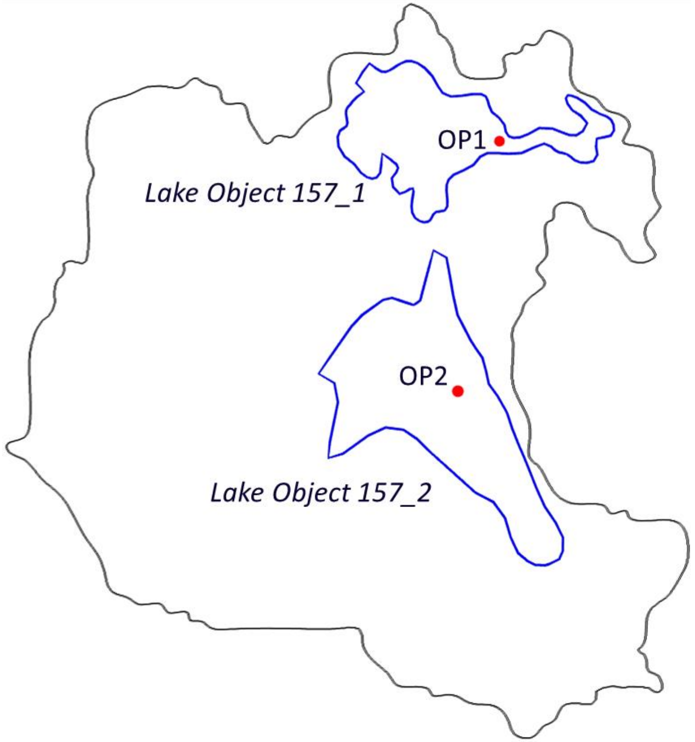
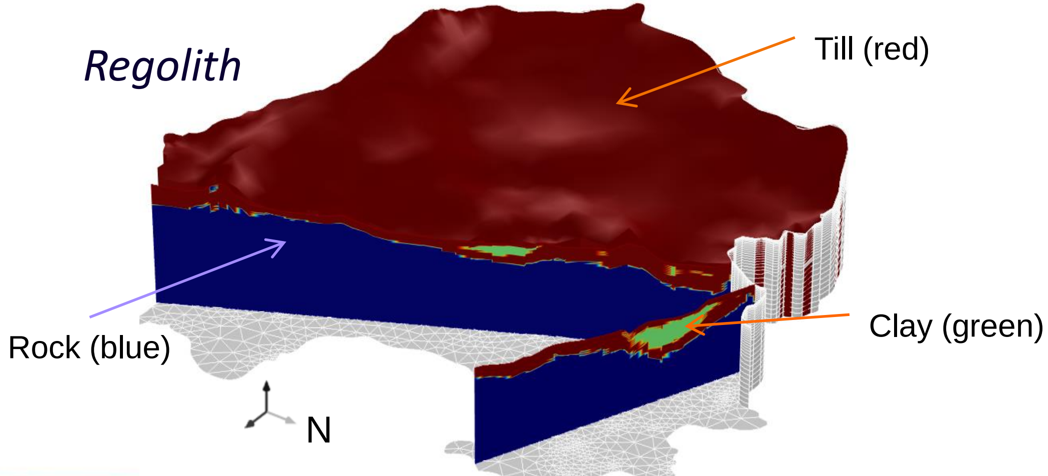


Figure 2-2. Conceptual model of the surface hydrology and near-surface hydrogeology at Forsmark.

Radionuclide (RN) sorption is often simulated using a lumped approach where retention processes are represented by the **distribution coefficient (K_d)**, which relates the radionuclide mass retained in the solid phase to its aqueous concentration.

K_d-based simulations rely on two strong assumptions: K_d depends on soil properties and is constant in time. However, sorption processes depend also on the fluid characteristics, which can vary in space and time.





How will be the evolution of RN at the near-surface system at Forsmark?

AMPHOS 21 — Scientific and Strategic Environmental Consulting

1994-2014 20 YEARS FLOWING

Objectives

The main objective of this work is to develop equivalent K_d-based radionuclides (RN) transport models that can reproduce the hydro-geochemical evolution of the soils at the site for spent nuclear fuel (Forsmark, Sweden), and evaluate their capabilities and limitations.

The specific objectives are

- To implement a 3D mechanistic reactive transport model of the regolith in iCP accounting for the evolution of ^{90}Sr, ^{137}Cs, ^{235}U and Ra.

- To use reactive transport model results to derive equivalent K_d values and/or distributions that are representative of the actual geochemical conditions of the regolith.

- Implement these functions into a K_d-based transport model in Comsol and compare the simulated retention evolution of RN with that obtained with the fully reactive transport model implemented in iCP.

Fully reactive transport model – iCP (Comsol-PhreeqC)

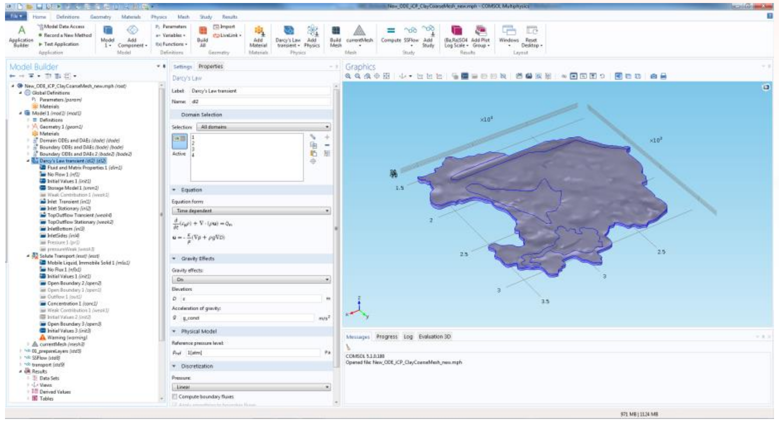
The coupled system of equations is solved using the widely spread Sequential Non Iterative Approach (SNIA), which is based on the Operator Splitting concept.

Flow in porous media

$$\frac{\partial(\phi\rho_l)}{\partial t} = \nabla \cdot \rho_l \boldsymbol{q}_l$$

Porosity updated from mineral precipitation

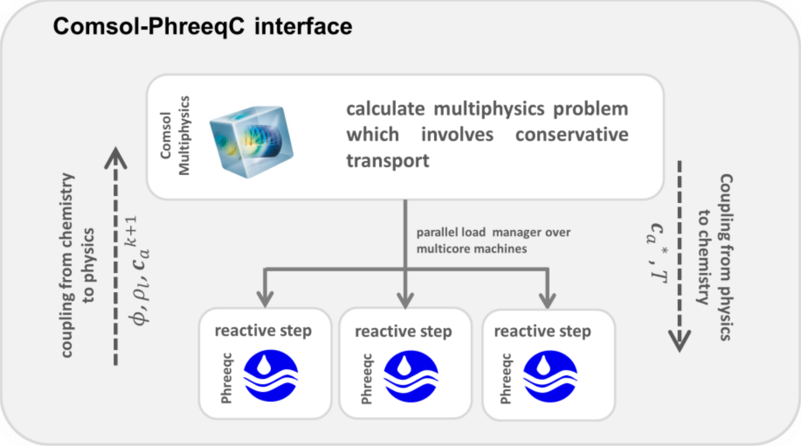
$$\phi^{k+1} = \phi^{k} + \omega_{\mathrm{l}}^{\mathrm{w}} \sum_{m=1}^{N_m} V_m \, \mathrm{c}_m^{\mathrm{k+1}}$$



Reactive transport

$$\boldsymbol{U}_a \frac{\partial(\phi\rho_l \boldsymbol{c}_a)}{\partial t} + \boldsymbol{U}_d \frac{\partial(\phi\rho_l \boldsymbol{c}_d)}{\partial t} + \boldsymbol{U}_m \frac{\partial((1-\phi)\rho_m \boldsymbol{c}_m)}{\partial t} = \boldsymbol{U}_a L_l(\boldsymbol{c}_a) + \boldsymbol{U}\boldsymbol{S}_{\boldsymbol{k}}{}^{\boldsymbol{t}} \boldsymbol{r}_{\boldsymbol{m}}(\boldsymbol{c})$$

Interface iCP



The interface Comsol-PhreeqC (iCP) was used to perform the fully reactive transport simulations. iCP combines the key capabilities of PhreeqC and Comsol in a single reactive transport simulator.

Retention processes

The radionuclides (^{90}Sr, ^{137}Cs, ^{235}U, Ra) are retained in the system according to the following processes

Radionuclide	Retention processes
^{90}Sr	- Adsorption in planar sites - Precipitation of Strontianite
^{137}Cs	- Adsorption in planar sites - Adsorption in type II sites - Adsorption in X_{FES} sites
^{235}U	- Adsorption on $Fe(OH)_3$ surface (strong and weak sites) (only in the Till) - Precipitation as Uraninite (only in the Clay)
Ra	- Precipitation as Radiobarite

Linear sorption model - Comsol

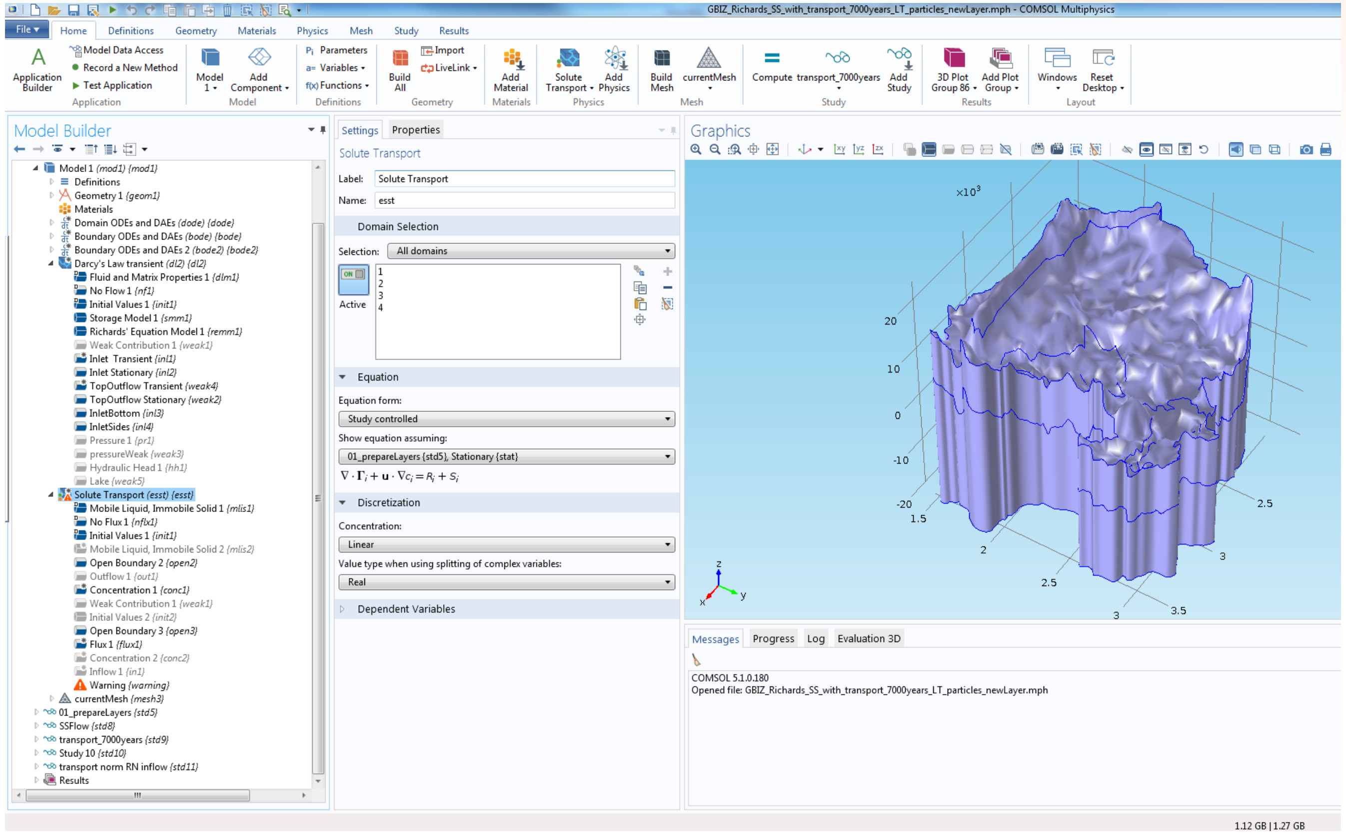
The K_d models were implemented in Comsol using the Darcy's Law (Subsurface Flow module, Fluid Flow), and Solute Transport (Chemical Species Transport module) physics and accounts for steady state saturated-unsaturated flow assuming constant recharge and transient RN transport with linear sorption.

Unsaturated groundwater flow

$$\rho\left(\frac{C_m}{\rho g}+S_e S\right)\frac{\partial p}{\partial t}+\nabla\cdot\rho\left(-\frac{k_s}{\mu}k_r(\nabla p+\rho g\nabla D)\right)=Q_m$$

Solute transport with linear sorption

$$(\theta_s+\rho_b k_{P,i})\frac{\partial c_i}{\partial t}+(c_i-\rho_P c_{P,i})\frac{\partial \theta_s}{\partial t}+\nabla\cdot(c_i\boldsymbol{u})$$
$$=\nabla\cdot\left[(D_{D,i}+\theta\tau_{L,i}D_{L,i})\nabla c_i\right]+R_i+S_i$$



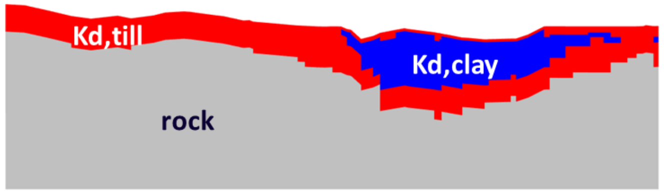
AMPHOS 21
SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

1994-2014
20 YEARS FLOWING

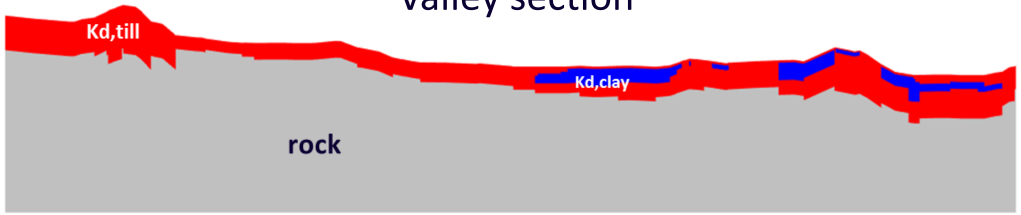
Geometry and mesh

Geometry: Digital elevation model (DEM) was imported as a parametric surface and extruded downward to generate the 3D geometry. The bottom of the model is intersected with a horizontal plane at z=-20 m

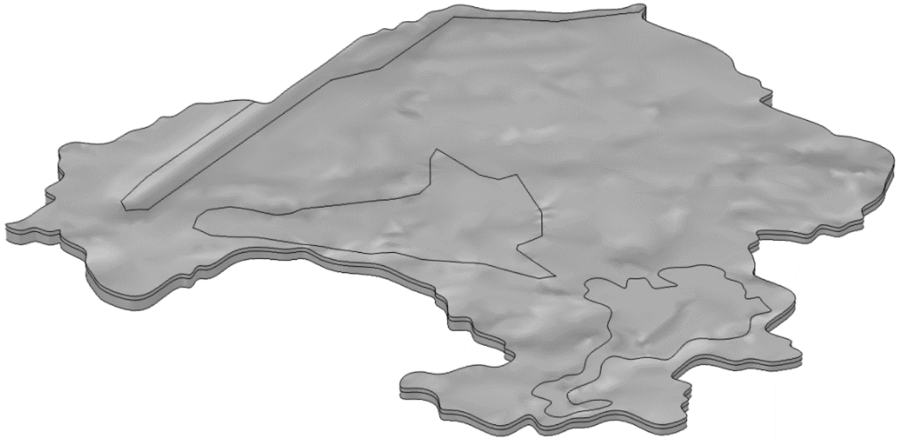
Lake section

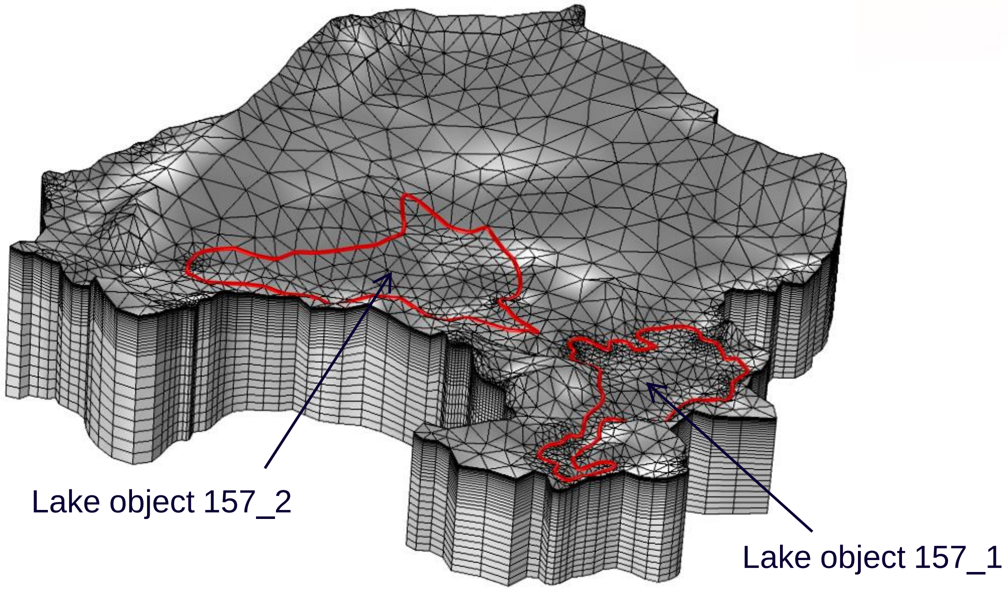


Valley section



Only two K_d zones are considered: till and clay (no retention in the rock).





The mesh is constituted by 85,380 prism elements of linear order. The element size is smaller in near the discharge areas

AMPHOS 21
Scientific and strategic environmental consulting

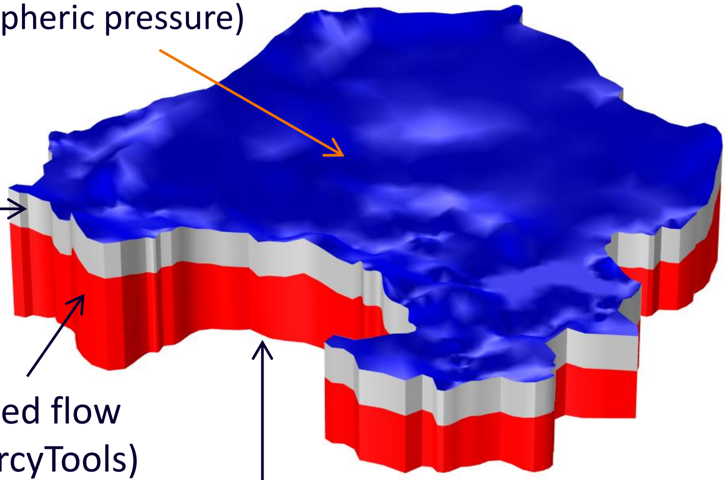
1994-2014 20 YEARS FLOWING

Boundary conditions

Groundwater flow

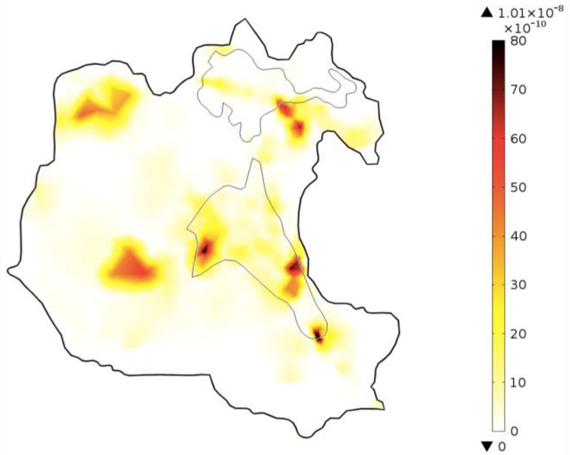
Prescribed recharge (infiltration fluxes from MIKE SHE) and discharge (atmospheric pressure)

No flow



Prescribed flow (from DarcyTools)

Prescribed inflow at the bottom (z = -20 m)

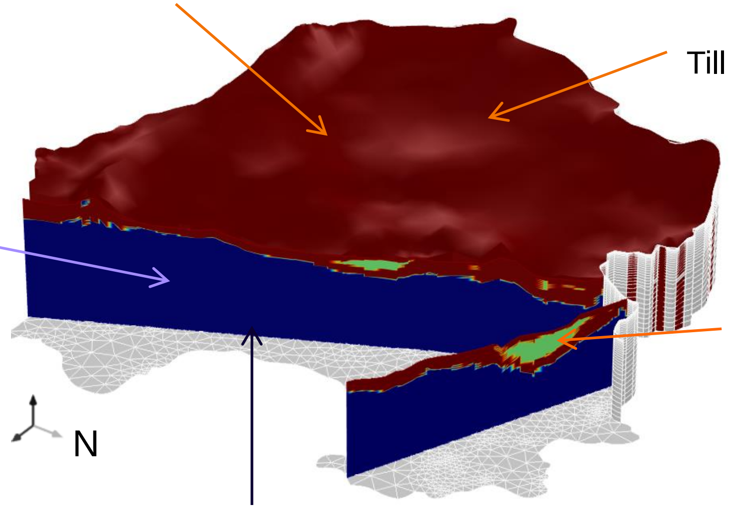


Solute transport

Top and lateral: Open boundary

Till (red)

Rock (blue)



Clay (green)

Bottom: Dirichlet BC

Concentrations in mol/kgw

RN	Clay, initial	Till, initial	Rock, initial	Bottom, Dirichlet	Top, Open Boundary
^{90}Sr	6.228×10^{-7}	2.098×10^{-6}	4.130×10^{-5}	8.781×10^{-4}	2.098×10^{-6}
^{137}Cs	4.503×10^{-11}	6.476×10^{-11}	1.162×10^{-8}	7.800×10^{-8}	6.476×10^{-11}
^{235}U	1.979×10^{-9}	2.234×10^{-8}	4.260×10^{-10}	1.7798×10^{-8}	2.234×10^{-8}
Ra	5.030×10^{-15}	5.030×10^{-15}	5.030×10^{-15}	5.030×10^{-15}	5.030×10^{-15}

AMPHOS 21
SCIENTIFIC AND STRATEGIC ENVIRONMENTAL CONSULTING

1994-2014 20 YEARS FLOWING

Equivalent K_d approaches

The results from a mechanistic reactive transport model (iCP) were used to calculate equivalent K_d fields for all the radionuclides modelled (Ra, Cs, U, Sr). Two types of equivalent K_d calculations were tested

(i) K_d spatial field (end of the iCP simulation)

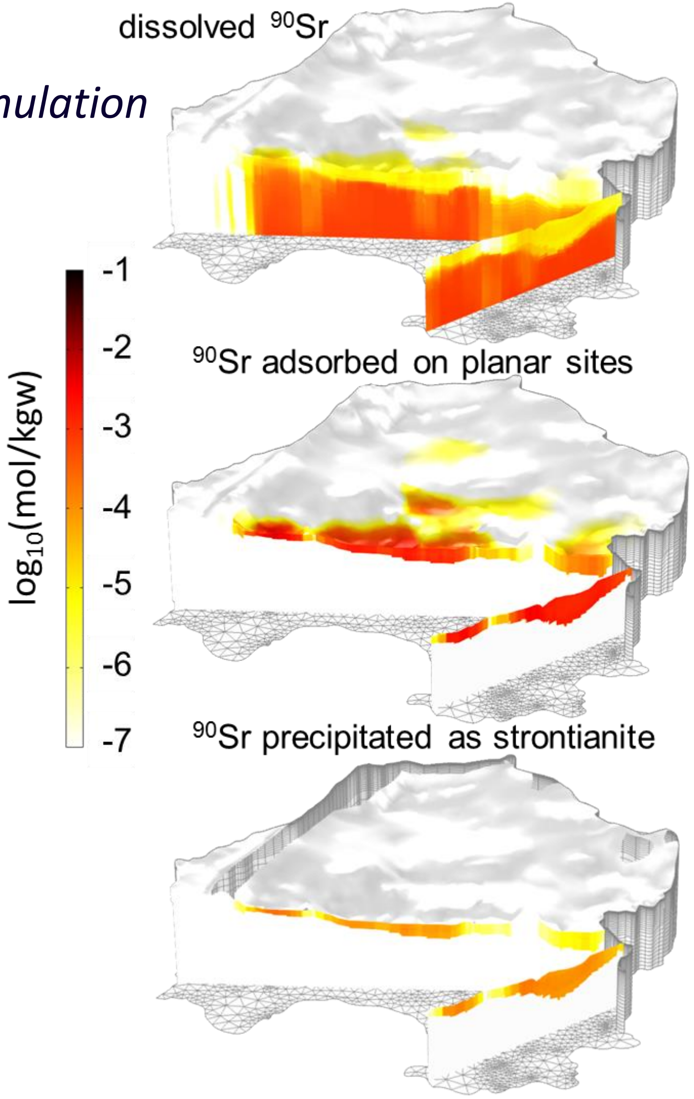
$$K_{d,RN}(x,y,z)=\frac{\theta_l(x,y,z)}{\rho_b}\frac{C_{RN,retained}(x,y,z)}{C_{RN,dissolved}(x,y,z)}$$

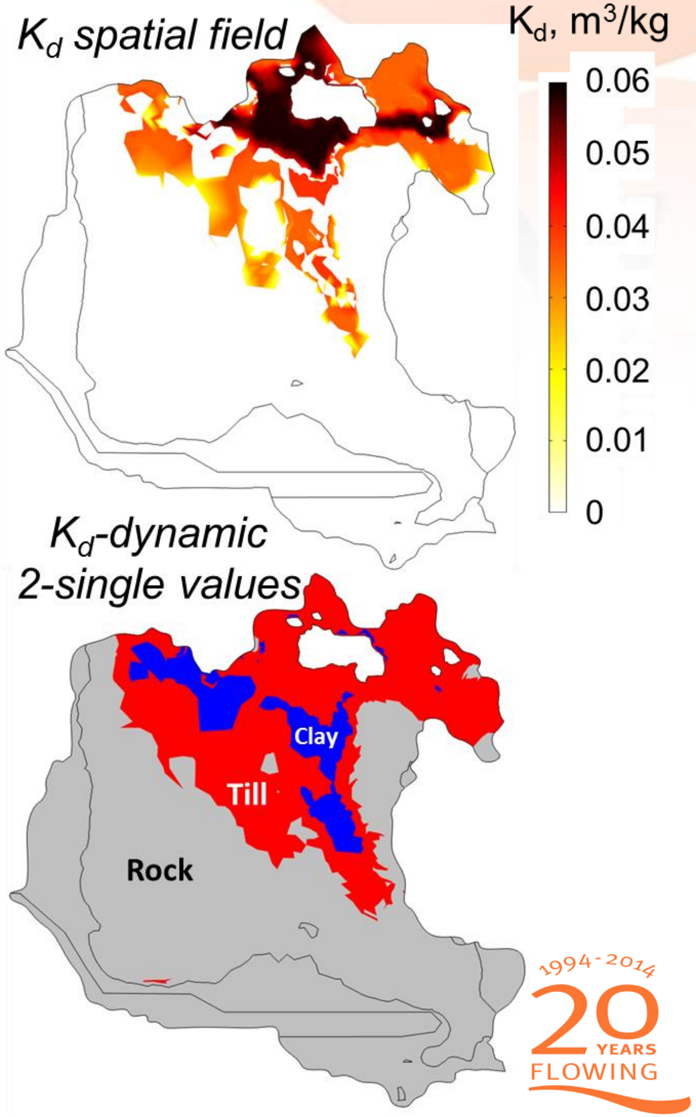
(ii) Dynamic 2-single K_d values: till and clay

$$K_{d,RN,\Omega}(t)=\frac{1}{V_\Omega}\left(\int_\Omega \frac{\theta_l(x,y,z)}{\rho_b}\frac{C_{RN,retained}(x,y,z)}{C_{RN,dissolved}(x,y,z)}\right)(t)$$

θ_l is the volumetric liquid content, ρ_b is the soil matrix density; Ω represents a layer (Till or Clay); V_Ω is the volume of layer Ω

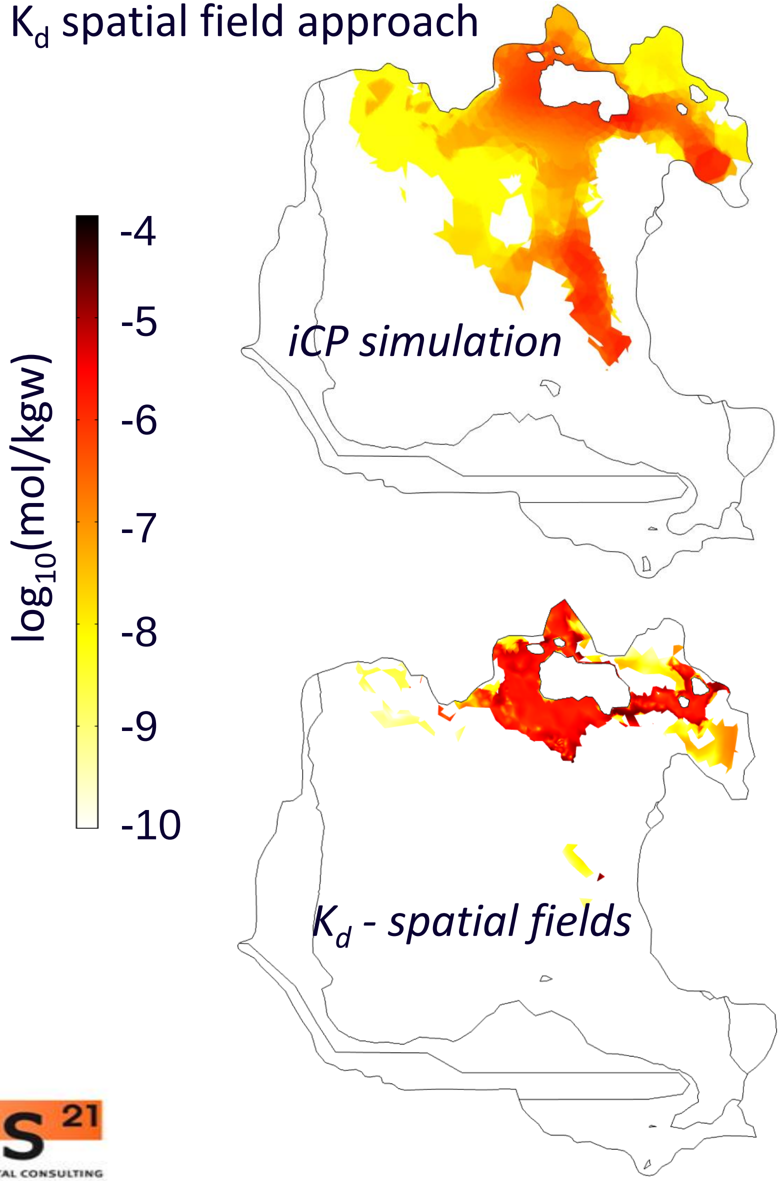
iCP simulation

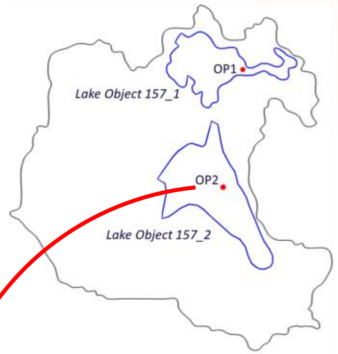


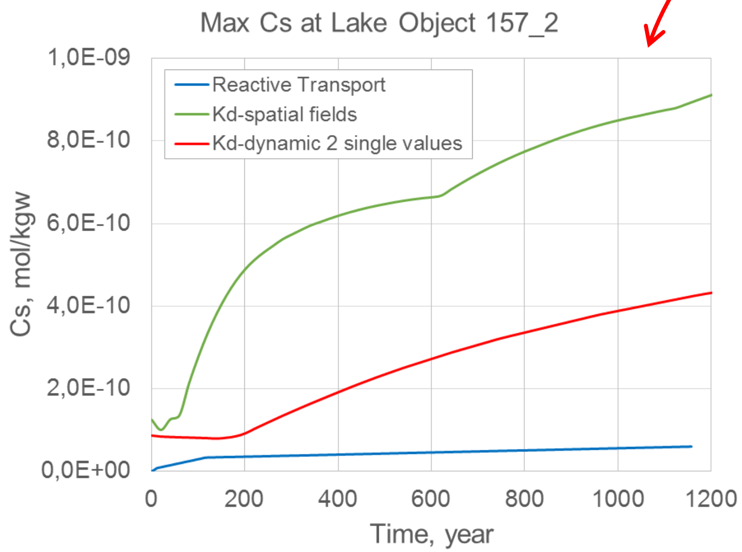


Results – Sorption of ^{167}Cs (z= -3 m, 1100 years)

The ^{137}Cs evolution suggests that a dynamic 2-single K_d values approach is better than the equivalent K_d spatial field approach



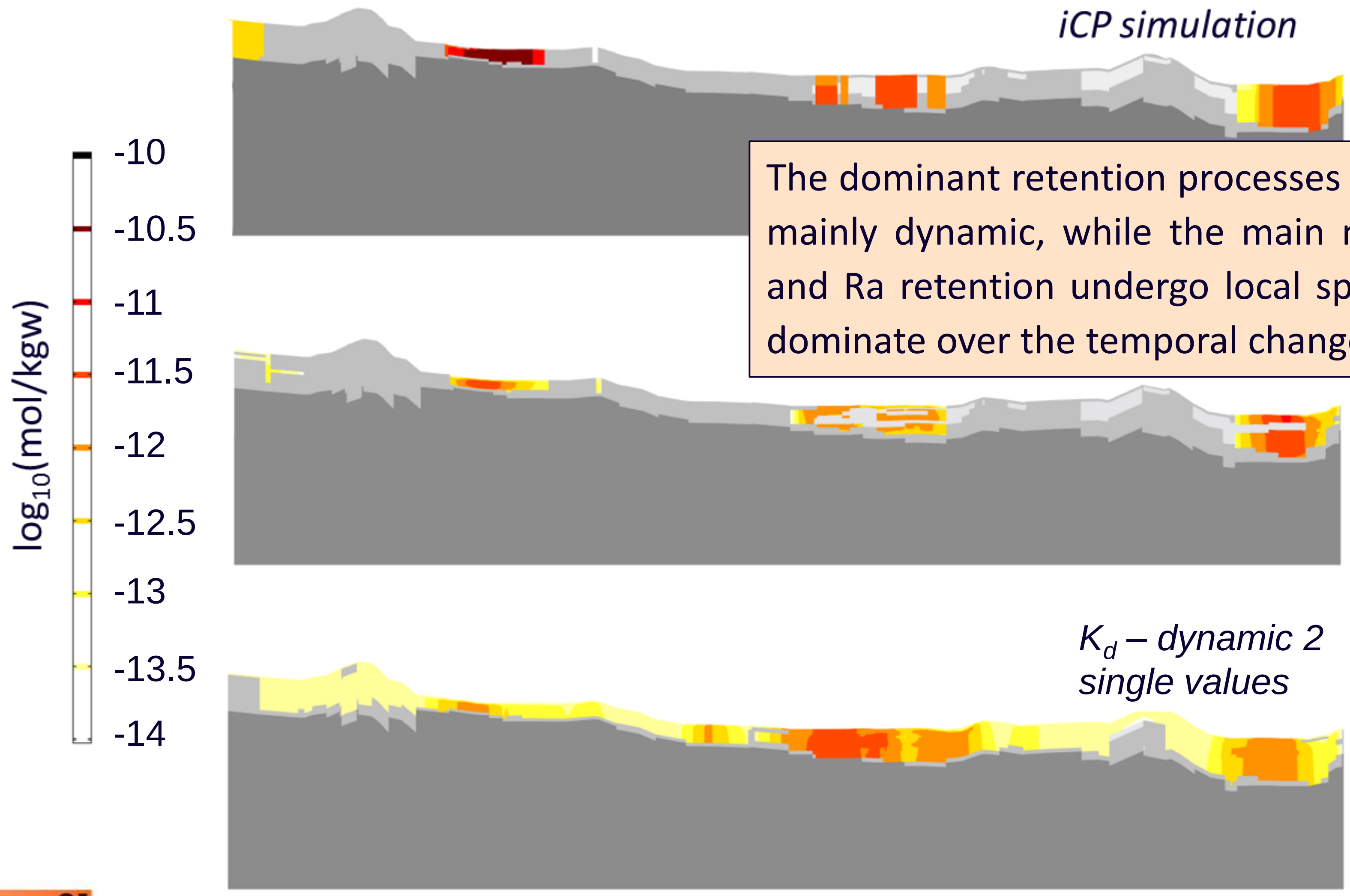




Breakthrough curves show discrepancies between both equivalent K_d-based transport models and the fully reactive transport model.

Results – Sorption of Ra (valley cross section, 1100 years)

The K_d spatial field approach reproduces ^{235}U and Ra retention better than the dynamic 2-single K_d values.



The dominant retention processes of ^{90}Sr and ^{137}Cs are mainly dynamic, while the main mechanisms of ^{235}U and Ra retention undergo local spatial variations that dominate over the temporal changes.

Conclusions

- Two equivalent K_d-based models were implemented in Comsol using the data calculated with the reactive transport model implemented in iCP: "K_d spatial field" and "dynamic 2-single K_d values".

- The formulation based on dynamic 2-single K_d values reproduces ^{90}Sr and ^{137}Cs retention better than the equivalent K_d spatial field approach, while this reproduces ^{235}U and Ra retention better than the former. This is because the dominant retention processes of ^{90}Sr and ^{137}Cs are mainly dynamic, while the main mechanisms of ^{235}U and Ra retention undergo local spatial variations that dominate over the temporal changes.

- The equivalent K_d-transport models presented in this work may capture a big portion of the regolith evolution if the main processes governing its chemical behaviour are interpreted properly.

- These models could be improved by approaches accounting for a dynamic update of the K_d values or by other alternative non-local in time and space approximations.

ESPAÑA
AMPHOS 21 CONSULTING S.L.
Paseo de García Faria, 49-51
08019 BARCELONA
Tel.: +34 93 583 05 00
Fax : +34 93 307 59 28

CHILE
AMPHOS 21 CONSULTING CHILE Ltda.
Av. Nueva Tajamar 481 of. 1005 (Torre Sur)
Las Condes 7550099
SANTIAGO DE CHILE
Tel.: +562 27991630

PERÚ
AMPHOS 21 CONSULTING PERU S.A.C.
Av. del Parque Sur 661, San Borja
Lima 41
Tel.: +511 5921275

FRANCE
AMPHOS 21 CONSULTING FRANCE S.A.R.L.
14 Avenue de l'Opéra
75001 PARIS
Tel.: +33 1 69345030

1994-2014 20 YEARS FLOWING

www.amphos21.com