

A COUPLED HYDRO-BIO-GEOCHEMICAL REGIONAL MODEL FOR THE ANALYSIS OF THE PERTURBATIONS CAUSED BY THE CONSTRUCTION OF A HLW REPOSITORY IN CRISTALLINE ROCKS

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ABSTRACT

We present a coupled hydro-bio-geochemical model to evaluate the transient perturbations caused by the construction of the network of galleries of a HLW repository in crystalline rock. A 2-D finite element model inspired on the numerical model of the Redox Zone experiment performed at the Äspö site (Sweden) is used to evaluate to what extent the conclusions of such experiment, which was performed at a depth of 70 m and lasted for a few years, can be extrapolated to the conditions of the actual repository located at 500 m depth and with galleries possibly open for some lustra. The model has been run under two hypotheses: 1) Abiotic model which considers only hydrochemical processes and 2) Biotic model in which both geochemical and microbial processes are taken into account. While conservative species shows dilution trends associated with the arrival of shallow fresh waters, concentrations of reactive species such as Fe^{2+} , bicarbonate and sulphate show a marked increase induced by microbial processes. The results of the current model show similar trends to those of the Redox Zone Model. There are some differences, however, due to differences in the tunnel depth and hydrodynamic parameters. While the Redox Zone experiment took place in a vertical fracture zone, the model of the repository applies to the bulk host rock which is less permeable. Model results are extremely sensitive to the specific growth rate of yeast, the half-saturation constant of POC, the initial concentration of POC in the upper layer and the distribution coefficient of POC. The model lacks sensitivity to the specific growth rate of iron reducer, the half-saturation constant of DOC and the yield coefficients of DOC to iron-reducer and that of POC to yeast.

1. INTRODUCTION

Fractured granite has been selected by several countries as a candidate host rock for a Deep Geological Repository (DGR) for high-level radioactive waste. Water flow and solute transport through fractured granite is a key factor for the design of a DGR because migration through the hosting granite would be the pathway of radionuclides to the biosphere. The assessment of the safety of such repositories requires using numerical models of groundwater flow, solute transport and chemical processes. These models are being developed from data and knowledge gained from in situ experiments such as the Redox Zone Experiment carried

out at the underground laboratory of Äspö in Sweden [Banwart et al., 1994; 1996; Samper et al., 2003; Molinero et al., 2004; Molinero and Samper, 2006].

Construction of the repository may perturb the hydrobiogeochemical conditions of the host rock. This paper presents a model which aims at evaluating the effect of the construction of a HLW repository on the hydro-biogeochemical conditions at the host rock. Special attention is paid to the role of microbial processes in restoring the prevailing reducing conditions. The model presented here is inspired in the model of the Redox Zone Experiment of Molinero (2000), Samper et al. (2003), Molinero et al. (2004) and Molinero and Samper (2006).

2. REACTIVE TRANSPORT MODELLING

2.1 Groundwater flow model

The repository will be constructed in a crystalline rock by the sea in an area where the idealized hydrogeological conditions are those illustrated in Figure 1. The repository is located at a depth of about 500 m and extends 1000 m in the horizontal direction. Flow is assumed to occur in a vertical plane within a rectangular domain 7000 m long and 2000 m deep with the repository located at 500 m depth. One can see in the Figure 2 that the domain is symmetric. Therefore, the model domain covers only a half of the full domain, that is, a rectangle 3500 m long and 2000 m deep. A groundwater recharge of 25 mm/year takes place at the top boundary. Under natural conditions, groundwater discharges to the sea. Figure 2 shows a contour plot of computed hydraulic heads and the velocity field under natural conditions.

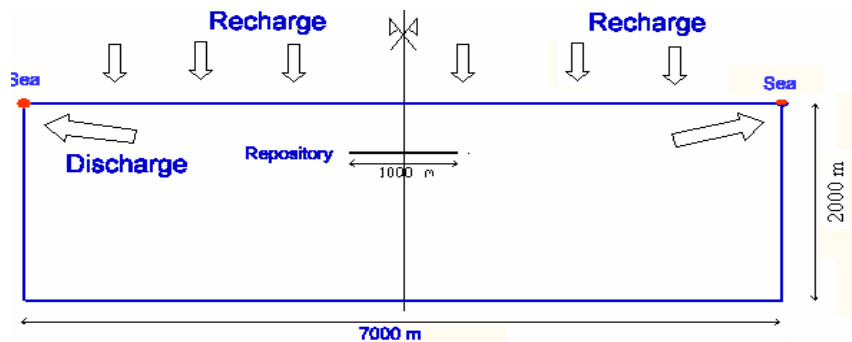


FIGURE 1. Sketch of the hydrogeological conceptual model.

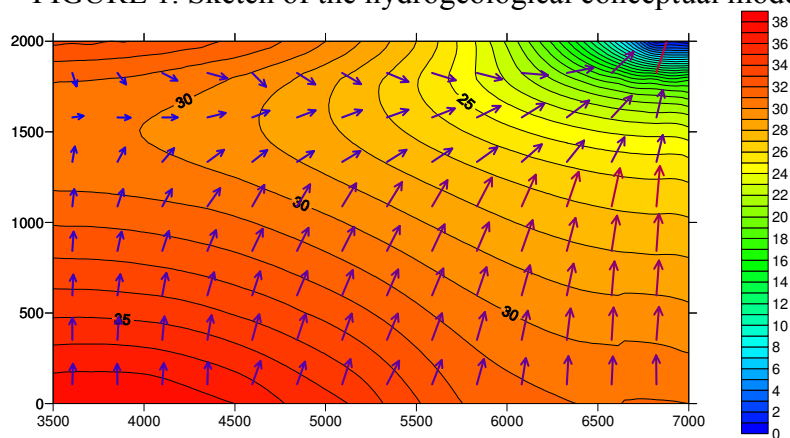


FIGURE 2. Groundwater flow field under natural conditions.

According to Rhén et al. (1997), the granitic rock at the Äspö site shows a vertical stratification. A layered zonation pattern in this study is adopted for hydraulic parameters which are similar to that of Jacquet and Siegel (2003) for simulating groundwater flow affected by glaciation at Äspö site. The domain consists of four stratified layers with 6 parameter zones. The hydraulic parameter values of each zone are listed in Table 1. Anisotropy of permeability was considered only for the upper layer with a ratio of 10. The storage coefficient is assumed equal to 10^{-8} and uniformly distributed in the whole domain.

The preferential flow due to the connected fractures is not taken into account in this study. Along the bottom boundary groundwater heads were fixed to values ranging from 34 to 40 m with a quasi linear distribution. A mixed boundary condition is prescribed at the sea with a leakage coefficient of $10 \text{ m}^2/\text{day}$. The repository was simulated with a mixed boundary condition. In order to simulate different stages (natural conditions, duration of construction of the repository and the repository closure) in a single run, a time function is used for leakage coefficients at the repository which are equal to zero under natural conditions, $10^{-6} \text{ m}^2/\text{day}$ during the phase of open repository (around 50 years) and zero after sealing the repository. No water flux was imposed on the remaining boundaries. Model domain was discretized with 365 nodes and 677 triangular finite elements (Figure 3). The grid is finer the repository and coarser near to the bottom.

TABLE 1. Main hydraulic parameters used in the 2-D model.

No.	Permeability (m/day)	Porosity	Longitudinal dispersivity (m)	Transverse dispersivity (m)
1	$7 \cdot 10^{-2}$	10^{-3}	100	60
2	10^{-4}	$5 \cdot 10^{-4}$	150	100
3	10^{-4}	$5 \cdot 10^{-4}$	150	100
4	10^{-5}	$1 \cdot 10^{-4}$	160	110
5	$2 \cdot 10^{-5}$	$1 \cdot 10^{-4}$	160	110
6	10^{-5}	$5 \cdot 10^{-5}$	300	150

2.2 Transport model

Porosity was assigned based on the values of permeability according to the functional expression proposed by Rhén et al. (1997). Longitudinal dispersivities range from 100 m in the upper layer to 300 m in the lowermost layer where the grid size is largest in order to ensure element Peclet number less than 2 to avoid numerical oscillations. Transverse dispersivity is about half of the longitudinal dispersivity (Table 1).

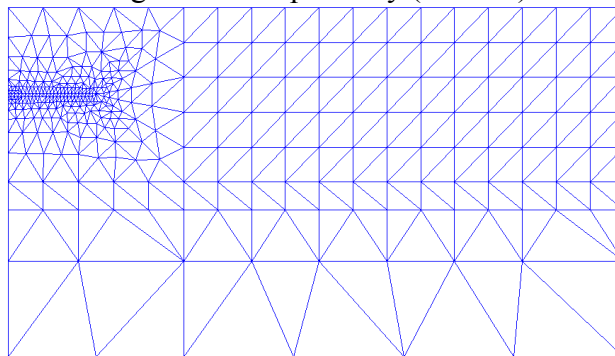


FIGURE 3. Finite element grid used for the hydrobiogeochemical model.

Aqueous concentrations defining the initial and boundary waters are provided in Table 2. The fresh water from the top boundary was obtained from the samples collected at the depth of 1-5 m. The saline water from the sea was obtained from the water samples collected from the sea floor sediments. The bottom boundary water was obtained the water samples collected from the deep granite at the Äspö site [Molinero and Samper, 2006].

The reactive transport calculations were performed with BIOCORE^{2D}, a code which has been extensively verified and successfully applied to laboratory and field cases [Zhang, 2001; Samper et al., 2003; Molinero et al., 2004; Samper et al., 2005].

2.3 Biogeochemical model

The biogeochemical model considered here is same as those of Samper et al. (2003) and Molinero et al. (2004) to interpret chemical data from the Redox zone experiment. Eighteen primary species and more than 60 additional aqueous complexes were selected to describe the aqueous solution in the chemical system. The equilibrium constants for aqueous speciation reactions and mineral solubilities were those from the EQ3NR database (Wolery, 1992). The composition of the host rock was described using the following minerals: calcite, hematite, pyrite, quartz and inert minerals which are not relevant to this study and therefore are not considered in the biogeochemical model.

TABLE 2. Total concentrations of solutes of the initial and boundary waters (mol/L).

	Initial	Boundary		
	saline water	fresh water	bottom water	sea water
Mg ⁺²	8.10×10 ⁻³	1.64 ×10 ⁻⁴	8.10×10 ⁻³	5.43×10 ⁻³
pH (-)	7.5	7.52	7.5	7.5
HCO ₃ ⁻	1.05×10 ⁻³	2.24 ×10 ⁻³	1.05×10 ⁻³	1.47×10 ⁻³
O _{2(aq)}	1.48×10 ⁻⁶⁶	1.48×10 ⁻⁶⁶	1.48×10 ⁻⁶⁶	1.48×10 ⁻⁶⁶
Cl ⁻	2.25×10 ⁻¹	1.69×10 ⁻⁴	2.25×10 ⁻¹	1.00×10 ⁻¹
K ⁺	4.50×10 ⁻⁴	6.65×10 ⁻⁵	4.50×10 ⁻⁴	2.33×10 ⁻⁴
Ca ⁺²	4.96×10 ⁻²	9.58×10 ⁻⁴	4.96×10 ⁻²	2.43×10 ⁻³
Fe ⁺²	1.13×10 ⁻⁵	1.00×10 ⁻⁵	1.13×10 ⁻⁵	1.13×10 ⁻⁵
SO ₄ ⁻²	4.95×10 ⁻³	2.50×10 ⁻⁴	4.95×10 ⁻³	2.60×10 ⁻²
Br ⁻	3.78×10 ⁻⁴	1.00×10 ⁻⁴	3.78×10 ⁻⁴	3.78×10 ⁻⁴
SiO _{2(aq)}	9.32×10 ⁻⁵	9.98×10 ⁻⁵	9.32×10 ⁻⁵	9.32×10 ⁻⁵
Mn ⁺²	1.71×10 ⁻⁵	6.55×10 ⁻⁶	1.71×10 ⁻⁵	1.71×10 ⁻⁵
Li ⁺	8.00×10 ⁻⁵	1.44×10 ⁻⁶	8.00×10 ⁻⁵	4.61×10 ⁻⁵
Na ⁺	1.09×10 ⁻¹	6.69×10 ⁻⁴	1.09×10 ⁻¹	8.52×10 ⁻²
Sr ⁺²	2.45×10 ⁻⁴	2.51×10 ⁻⁶	2.45×10 ⁻⁴	2.45×10 ⁻⁴
DOC(g/L)	1.00×10 ⁻⁴	3.00×10 ⁻²	1.00×10 ⁻⁴	1.00×10 ⁻⁴
POC(g/L)	1.00×10 ⁻⁸	3.00×10 ⁻²	1.00×10 ⁻⁸	1.00×10 ⁻⁸
Fe_reducer(mg/L)	1.058×10 ⁻⁶	1.058×10 ⁻⁶	1.058×10 ⁻⁸	1.058×10 ⁻⁸
Yeast (mg/L)	1.058×10 ⁻⁶	1.058×10 ⁻⁶	1.058×10 ⁻⁸	1.058×10 ⁻⁸

Two microbial processes were taken into account in this study 1) fermentation of POC by yeasts at the top layer and 2) degradation of DOC by iron-reducers. POC is assumed to be an aqueous species rather than a species attached to the solid phase. Since POC has a low mobility, a large retardation coefficient is used to hinder the mobility of POC. The growth of both yeast and iron-reducer is governed by a single Monod kinetic rate [Samper et al., 2003].

The biogeochemical model was calibrated with the data measured at depth of 70 m from the Redox zone experiment. The model not only reproduces the overall trends of conservative and reactive species caused by dilution but also gives quantitative explanation of increase in concentrations of bicarbonate and sulphate at depth of 70 after an access tunnel intersects the fracture zone. Microbial kinetic parameters used in this study are adopted from those used in the model for the Redox zone experiment [Samper et al., 2003].

3. RESULTS

3.1 Natural conditions

Groundwater recharging along the upper boundary discharges into the sea at the right-upper corner of the domain (Figure 1). There is a small amount of flux from the bottom boundary upwards which is, however, less than 1% of the total discharge. Under natural conditions, the total discharge rate into the sea is equal to $0.022 \text{ m}^3/\text{day}$.

Figure 4 shows the spatial distribution of Cl^- concentration with a clear interface between fresh and saline water under natural conditions which is located at depth of about 200-300 m. Spatial distributions of other reactive species also show a clear stable interface between fresh and saline waters which is located above the repository. Those figures are not shown here.

3.2 Open repository

Construction of the repository is assumed to last up to 50 years. As long as the repository remains open, it acts as a hydraulic sink (Figure 5). Ground water discharge into the repository causes the interface between saline and fresh waters moving downward to the repository depth. Fresh water coming from the top layers leads to the dilution for most chemical species at the repository depth. Two hypotheses were considered: with and without microbial processes. In the case of accounting for microbial processes, concentrations of bicarbonate and sulphate at the repository depth significantly increases with time while in the case of without accounting for microbial processes concentrations of bicarbonate and sulphate slightly increase, the effect of dilution on the two species is counteracted by minerals dissolution. However, pH remains quite constant showing only a mild change at the depth of repository. The results are similar to those of Redox zone experiment model [Samper et al., 2003]. However, there are still several differences because the model of the Redox Zone experiment corresponds to a vertical fracture zone. In addition, the depth of the tunnel at the Redox Zone (70 m) is much smaller than that of the repository (500 m) in current model.

3.3 Post-closure stage

After sealing the repository, the repository will no longer act as a hydraulic sink. Figure 5 also shows the spatial distributions of water heads computed at 10, 50, 100 and 450 years after the sealing of the repository. One can see that groundwater discharges only into the sea and the flow field slowly recovers to that under natural conditions. The time for groundwater flow needed to recover to that under nature conditions is directly proportional to L^2 (the depth

of the repository) and inversely proportional to the hydraulic diffusivity (ratio of permeability and storativity). According to the parameter values used in the model, 600 years are obtained after the sealing of the repository [Samper et al., 2005].

Figure 4 shows also spatial distributions of Cl^- after the sealing of the repository. One can see that even at 1750 years after the sealing of the repository spatial distribution of Cl^- can not recover to that under natural conditions. It is very interesting that the interface between saline and fresh water keeps moving slowly downward during 250 years since the sealing of the repository. After that time, the interface moves slowly upwards. Extrapolating the trend of computed concentrations of Cl^- at the depth of the repository from the semilog plot, it is estimated that almost 8000 years are needed to recover to the concentrations under natural conditions.

After sealing of the repository, bicarbonate and sulphate keep increasing almost 200 years and later decrease. Figure 6 shows time evolutions of bicarbonate and sulphate at the repository depth after sealing the repository.

At the repository, pH slightly decreases from 7 to 6.5 during the open repository stage and remains constant for 1000 years. The trend of pE is reverse to that of pH because pE increases from -3 to -2 during the open repository stage, remains constant for 1000 years and starts to increase after 1800 years.

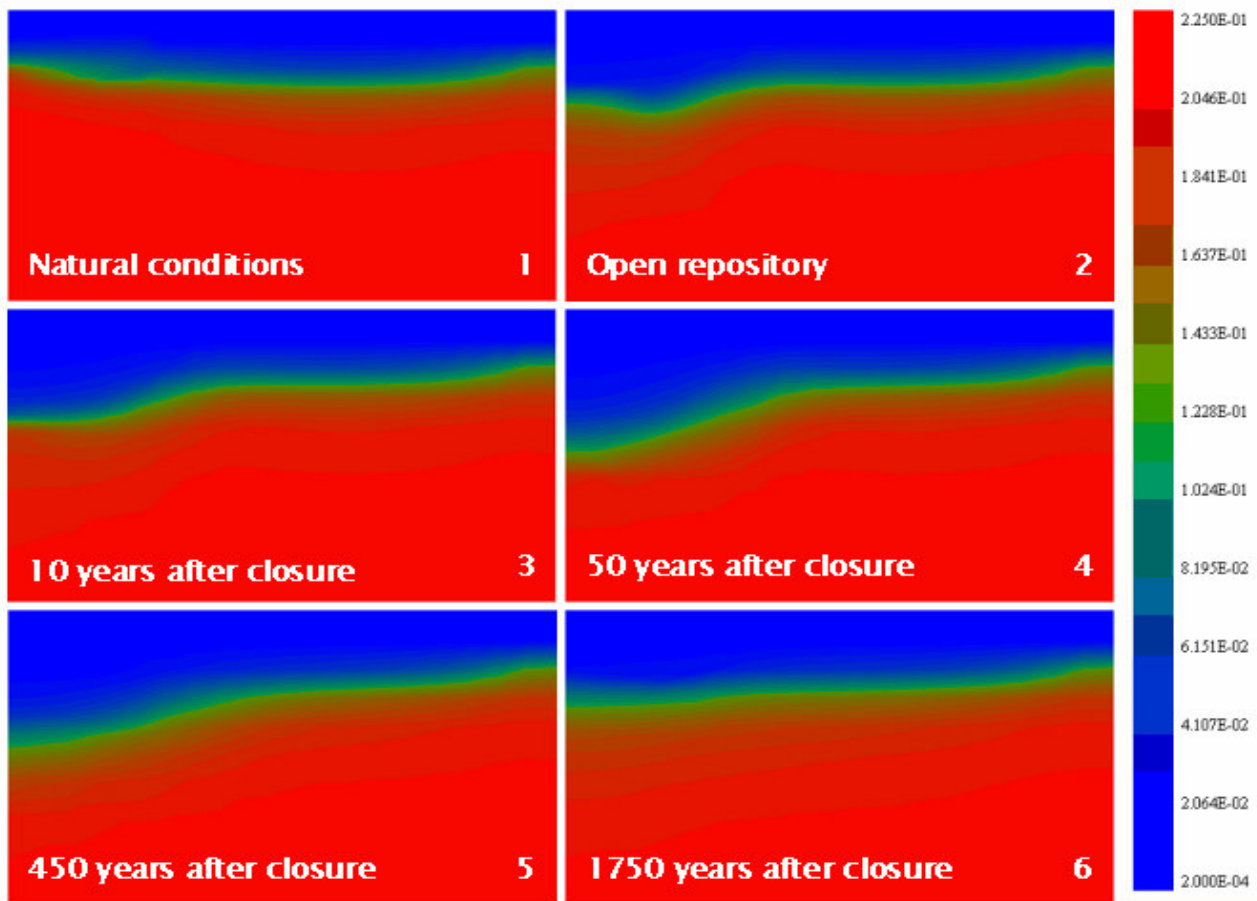


FIGURE 4. Spatial distribution of Cl^- at different time stages.

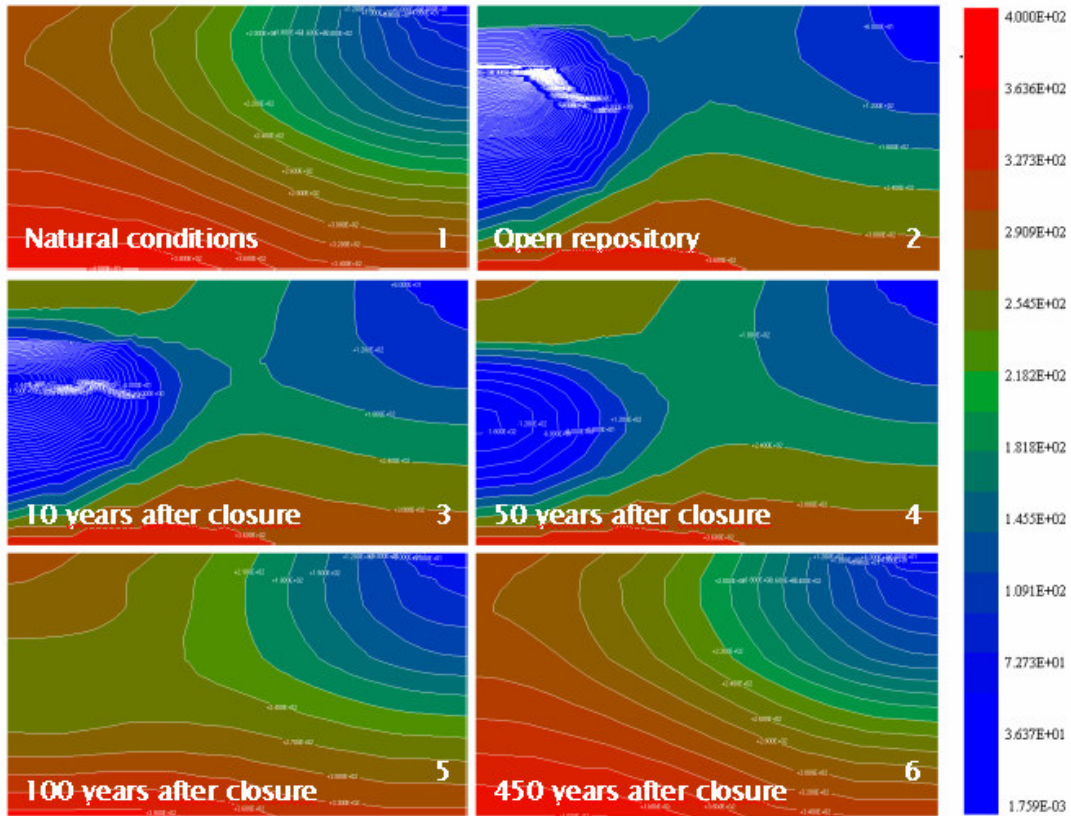


FIGURE 5. Spatial distribution of hydraulic heads at different time stages.

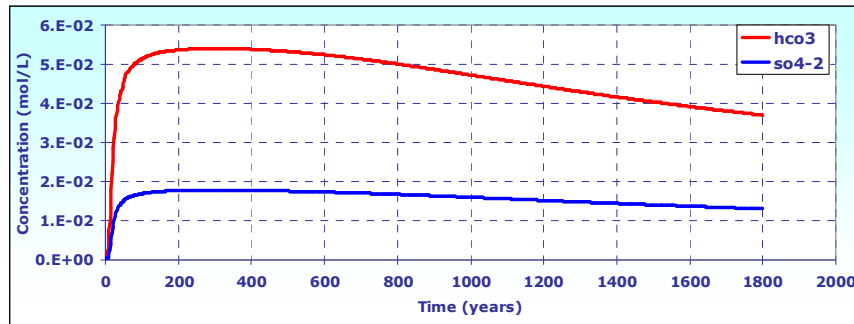


FIGURE 6. Long-term time evolution of the concentrations of HCO_3^- and SO_4^{2-} at the repository depth.

4. CONCLUSIONS

A coupled hydrobiogeochemical model has been presented to evaluate the perturbations caused by the construction of a HLW repository in crystalline rocks. The biogeochemical model was calibrated with the data from the Redox experiment. The results of the model indicate that there is a hydraulic transient stage after closure of the repository which has a duration of 450 years. The dynamics of solute transport are slower than those of groundwater flow. Chemical and microbial processes retard the propagation of the hydraulic perturbation. Computed results for Cl^- indicate that even 1750 years after repository closure, Cl^- concentrations will not have reached their natural values. The fresh/saline water interface

moves downwards due to the construction of the repository. After repository closure the interface keeps moving slowly downwards for at least 250 years. After that time, molecular diffusion, which is a much slower process than advection, causes a progressive upwards movement of the interface. The simulation was run for 1800 years and the final Cl⁻ concentration at the repository depth was still far from the natural value. Extrapolating the trend of computed values, it is estimated that natural chemical conditions for Cl⁻ will be reached at least 8000 years after closure. Bicarbonate and sulphate increase during the open repository stage, keep increasing for about 200 years and later decrease. Computed DOC values remain stable with a long term trend to decrease. The time evolution of the concentration of Fe-reducer at the repository depth shows an unexpected trend with a peak value of 0.002 mg/L after 300 years. pH decreases from 7 to 6.5 during the open repository stage and remains constant for 1000 years. It starts to decrease again after 1800 years. The trend of pE is reverse to that of pH because pE increases from -3 to -2 during the open repository stage, remains constant for 1000 years and starts to increase at the end of the simulation time. A longer run should be performed in order to achieve steady concentrations of all chemical and microbial species. Future studies should address also the explanation of the behaviour of Fe reducers during the post-closure transient stage.

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