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Chemistry-transport coupled modelling of the Äspö groundwater system (Sweden) since the last glaciation

W. Kloppmann, D. Thiéry, C. Kervévan, A. Bourguignon, P. Négrel, J. Casanova
BRGM, BP 6009, F-45060 Orléans Cedex 2, France

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ABSTRACT: Based on the specific coupled modelling approach developed at BRGM, a reaction-transport model of the Äspö groundwater system (Sweden) has been developed; it simulates a continuous scenario from 12.000 BP to the natural modern situation. Examples of the results (evolution of Na, ^{18}O, calcite) are presented. Na, Ca, Cl and stable isotopes are mainly related to mixing between the different mixing endmembers subsequently entering the system (glacial meltwater, Baltic Sea stages, meteoric water), calcite contents, pH and silica are determined by water rock interaction.

1 INTRODUCTION

The long term prediction of the geochemical evolution of groundwater bodies within potential host formations is of particular concern for site assessment in the field of nuclear waste disposal (Boulton et al. in press). Evaluating the impact of changes in climatic/hydrological surface conditions on the groundwater quality has to make use of a combined approach of flow and transport modelling taking into account chemical reactions. An adapted methodology of chemistry-transport coupled modelling based on Specific Chemical Simulators (SCS) has been developed in the last few years (Kervevan et al. 1998). It was applied to the site of Äspö (Sweden) for which a detailed site investigation yielded huge quantities of geological, hydrogeological and geochemical data from the crystalline basement up to depths of >1000 m (Laaksoharju et al., 1999a). The paleohydrological/hydrochemical evolution since the last glacial maximum is particularly complex due to the presence of deep seated brines, meltwater injection, several phases of immersion of Äspö island by the subsequent stages of the Baltic Sea and a final emergence of the site allowing mixing with meteoric waters at the surface (Laaksoharju et al. 1999a).

In the following, we describe, as an example for the application of the SCS approach, the attempt to reconstitute the geochemical and flow conditions at Äspö from the last deglaciation to the modern natural conditions prevailing before the construction of the underground laboratory.

Figure 1. Situation of the study site

2 FLOW AND TRANSPORT MODELLING

The modelled zone is a square of 8 km length and extends to a depth of 1600 m below the present day sea level. All geometric (elevation) and hydraulic data (3 components of conductivity, hydraulic head, salinity) were provided by U. Svensson (1997). A compromise had to be found between computing time, spatial and temporal resolution of the transient state flow modelling and the complexity of the geochemical model. The final configuration adopted is a horizontal irregular grid of 26 x 26 cells and a vertical grid of 11 layers from 3 to 300 m of thickness.

The hydrodynamic code used is the finite difference model MARTHE (Thiéry, 1993a, b) taking into account an unsaturated zone, density effects, direc tional hydraulic conductivity (in fractures) and multi-component transfer integrating advection and mechanical dispersion.

The initial concentrations in chemical species have been considered as constant in the whole domain depending only on whether a cell is considered as crossed by fractures or as composed of matrix only. The ^{18}O signature is -8.9‰ in the initial dense brine; -20.45‰ in the initial mixed brine and –32.0 ‰ in the initial glacial meltwater.

The sea level, the concentrations and the stable isotope composition of the Baltic varied considerably with time. The boundary conditions were accordingly adjusted every 1000 year period. Salinity and hydraulic head calculations were performed 320 times for 1000 years with a frequency decreasing from the beginning to the end of each 1000 year stress period. The hydrochemical calculations were performed every 1000 years. A test with twice as much time steps yielded no significant difference for the concentrations of aquatic species.

3 GEOCHEMICAL MODELLING

3.1 *Development of the "PAGEPA" Specific Chemical Simulator (SCS)*

The general approach chosen by BRGM is to design a dedicated geochemical code (so called "SCS") developed specifically for each particular application that only takes into account the relevant processes to be considered (Kervévan et al. 1998). One of the advantages of this approach is a significant improvement in computer efficiency that in turn allows relatively complex 3D coupled modelling.

This approach thus requires us to develop a new SCS for each new application. The combined use of ALLAN 3.1 and the NEPTUNIX 4 software package automatically generates the coded part of the simulators, as long as the processes involved in the physical system being modelled can be described by a set of algebraic and/or ordinary differential equations.

NEPTUNIX 4 is a general solver which automatically generates a FORTRAN code solving the selected set of equations. ALLAN 3.1 functionalities include graphical interface, description language, translator for NEPTUNIX 4, graphical assembling of models (including the generation of coupling equations), exploitation of the simulators and post-processing of the generated data.

The SCS developed for the Äspö site includes 24 aqueous reactions of 12 elements (H, C, O, Ca, Al, Si, Mg, Na, Cl, K, Fe, S_{ox}), the ^{18}O isotope, 7 precipitation/dissolution reactions (calcite, kaolinite, clinochlore, goethite, quartz, K-feldspar, anorthite) and two cation exchange reactions (K and Na versus Ca).

3.2 *Chemical boundary conditions*

All calculations are based on the reference waters defined by Laaksoharju et al. (1999b) as the groundwater samples closest to the hypothetic pure mixing endmembers. In order to characterise the geochemical system in terms of major species to be taken into account, we performed preliminary EQ 3/6 (Wollery, 1992) calculations of chemical speciation and saturation states of all the reference waters. The species whose relative concentration was less than 10^{-2} with respect to the major species were not considered in the SCS.

The different inputs of marine and continental waters into the system during the past 12,000 years were defined and taken into account in the coupled simulations. In order to finalize our selection of aqueous species actually taken into account in the SCS, EQ3/6 mixing calculations were also performed to simulate chemical compositions, speciation and saturation states of the different Baltic Sea stages.

The three predominant lithologies encountered in the four main boreholes and in the tunnel are the Äspö diorite, the Småland granite and the Fine-grained granite (Mazurek et al., 1995 ; Stanfors et al. 1999). Extensive studies characterise the mineralogy of the fracture fillings (Mazurek et al., 1995; Tullborg, 1989; Wikman et al., 1988 ; Landström et al., 1993). A selection of the occurring minerals, taking into account the constraints related to computing, was integrated into the SCS.

3.3 *Chemical evolution*

As a general observation we can state that certain parameters, the conservative species like Cl, but also Na and bivalent cations, reflect essentially the mixing processes in the course of the subsequent stages of glacial/meteoric/marine waters injection whereas others (Si, pH, mineral precipitation) are closely related to the geometry of the reservoir and in particular of major fractures.

Figure 2 shows, as example of the modelling results, the distribution of the Na concentrations of the $\delta^{18}O$ and of calcite for 11,000, 6000 and 1000 years BP. All block diagrams show E-W sections cutting through Äspö Island.

Na: The patterns of the Na distribution are, much like those of Ca, closely related to the chlorinity. The initial scenario at 12,000 BP represents the stratification of glacial meltwater over deep brines. The fluctuations between 12,000 and 10,000 BP can be attributed to density effects. The influence of Yoldia sea becomes visible at the 9000 BP stage by increasing salinity of the surface layers. Ancylus lake waters (8000 BP) dilute the superficial strata and the Litorina stage increases again the the concentration in the surface waters. From 5000 BP onward, the progressive invasion of the system by low salinity meteoric waters from the West is driven by the increasing hydraulic gradient due to lowering of the sea level. The Baltic 2 stage introduces a second low Na plume from 1000 BP to present. Low chlorinities of the modern Baltic Sea (since 1000 BP) and of meteoric water determine the present day conditions in the surface layers. We observe a residual local salinity high near the Äspö island where Baltic 1 (or Litorina sea) water is conserved between the intrusion of meteoric water and modern Baltic Sea water.

Calcite: According to the defined initial conditions, calcite is limited to major fractures at 12,000 BP. Calcite precipitation in the fissure porosity of the matrix starts from the surface during the Baltic ice lake stage (Fig. 2). During the whole evolution of the site, we observe a rather continuous progress of a horizontal “calcite precipitation front” moving from the surface to depth. Obviously, calcite precipitation is fairly independent from the subsequent marine/meteoric entries into the system and from the major ion concentration distribution. From 3000 BP to present, calcite contents in the surface layers reach values comparable with the initial concentrations defined for the major fractures.

Silica: Si concentrations do not vary significantly during the evolution of the site. This is only partly due to the fact that Si variations in the initial fluids entering the system are small. The crucial factor seems to be a buffering of the concentrations by mineral dissolution/precipitation (according to the chemical model, silica concentrations are controlled by quartz).

pH: The total range of pH variations is relatively restrained (around 1 pH unit). The pH is definitely not an indicator of mixing but it’s distribution is closely related to mineral equilibria. The pH distribution is, much like silica concentrations and calcite contents, determined by the geometry of major fractures. In the fractures we observe the highest values around 9, which is in the uppermost part of the range of measured field values.

$\delta^{18}O$: Initially, the system is invaded by isotopically extremely depleted glacial meltwater, which mixes at depth with the more enriched brines (Fig. 2). The pattern of isotopic contents till 10,000 BP remains rather constant and is very similar to the sodium/chloride distribution (Fig 2). Both tracers can in fact be regarded as conservative under the conditions prevailing in the system. The impact of the ^{18}O-enriched Yoldia sea changes profoundly the isotopic contents of the system and the influence of the Yoldia water reaches depths of >900 m. At 8000 BP, the isotopic composition of the system is homogenised up to the bottom of the block where slightly enriched brine remains. The impact of isotopically enriched Litorina sea ($\delta^{18}O = -5.5‰$) becomes predominant from 7000 to 5000 BP and the isotopic composition of the Baltic Sea is constant up to present day conditions around -6‰. Meteoric water which is depleted ($\delta^{18}O = -10.5‰$) with respect to Litorina seawater progressively invades the system from the continent. Present day conditions are somewhat similar to chloride distribution but contrasts of the isotopic composition are much lower than for chlorinity.

4 CONCLUSIONS

The coupled hydrodynamic-SCS modelling approach has been successfully applied to a complex setting of a fractured coastal aquifer influenced by glacial melt water, meteoric water and successive stages of brackish to saline seawater intrusions over 12,000 y. The number of chemical reactions taken into account (in both space and time) are at present mainly limited by the constraints of computing capacity. Some simplifications of the natural system that were adopted in the present study, as neglecting redox processes though they seem to play a particular important role in this site (Banwart, 1995), could be overcome in a near future considering the fast evolution of computer capacities.

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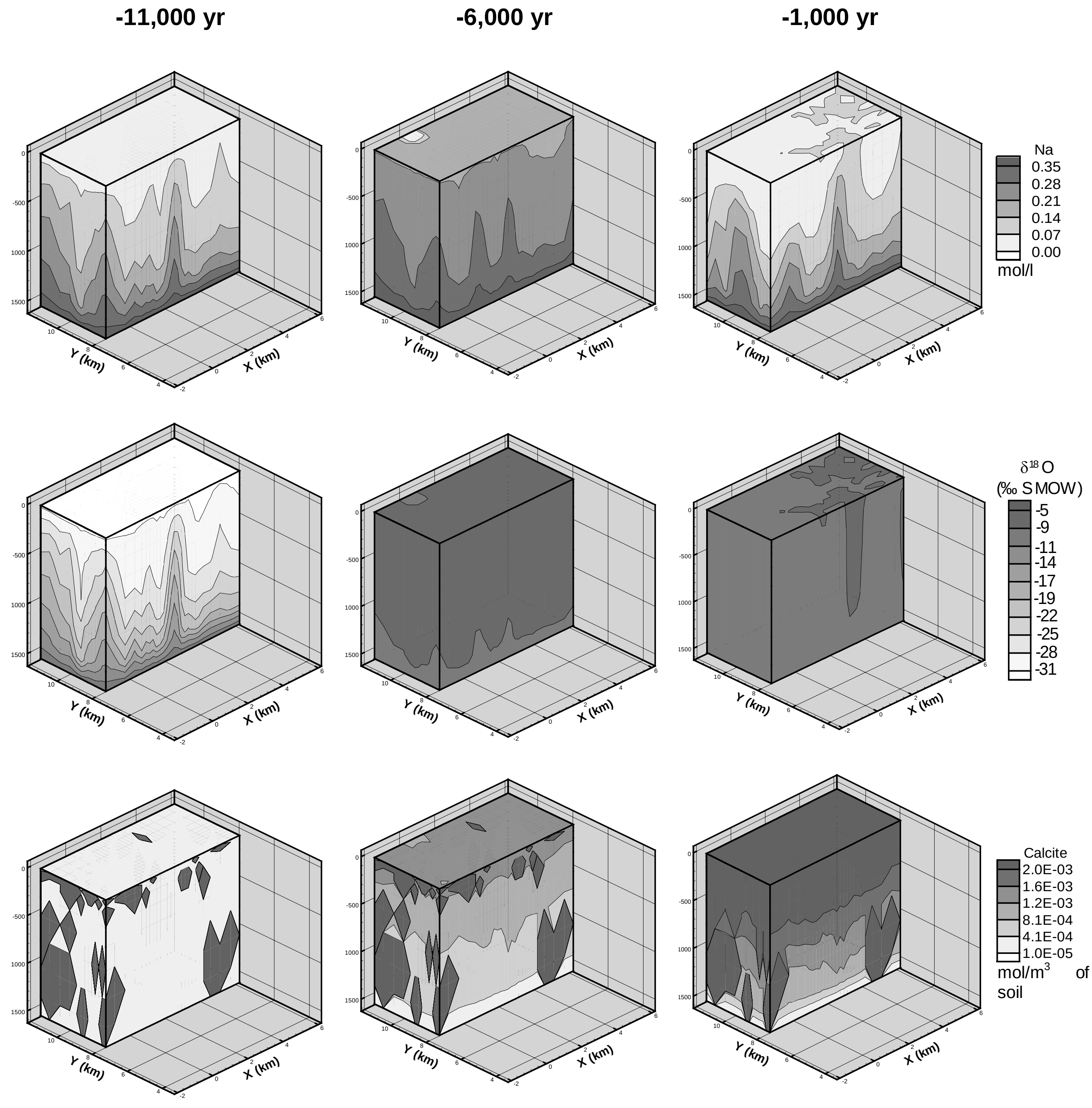

Figure 2. E-W section through the study area (lateral extension: 8 km x 8km, heith of the block: 1800 m): Distribution of solid calcite, dissolved Na and δ^{18}O at 11,000, 6000and 1000 BP.

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