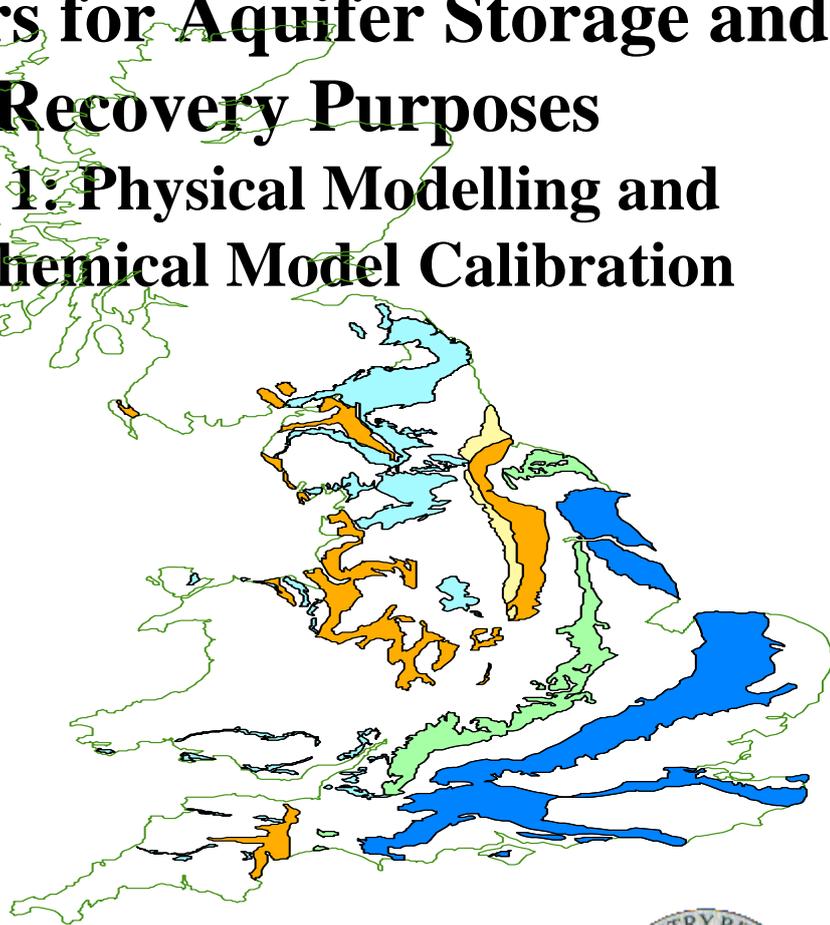


# ASR - UK

## Physical and Geochemical Modelling (SWIFT-PHREEQC) of British Aquifers for Aquifer Storage and Recovery Purposes

### Part 1: Physical Modelling and Geochemical Model Calibration



**BRITISH GEOLOGICAL SURVEY**  
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**Aquifer Storage and Recovery**

**Physical and Geochemical Modelling (SWIFT-  
PHREEQC) of British Aquifers for Aquifer Storage  
and Recovery Purposes**

**Part 1: Physical Modelling and Geochemical Model  
Calibration**

I Gaus, A T Williams & P Shand

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## Summary

This report describes the progress that has been made in developing models simulating both the physical and geochemical aspects of Aquifer Storage Recovery (ASR) schemes. This work is part of a 30 month project, entitled ASR-UK, which started in April 1999. The report describes progress to the end of 1999.

SWIFT is a fully transient 3-Dimensional model that simulates flow and transport of fluids. It is ideally suited to modelling ASR and has been used to simulate the response of the four major aquifers in UK – Chalk, Lincolnshire Limestone, Sherwood Sandstone and Lower Greensand.

Ranges of parameters applicable to these aquifers, which encompass dual-porosity, fractured and porous aquifers, were used to predict their response to a hypothetical, but realistic, cycle testing programme and an annual operational cycle over five years.

Sensitivity analyses of the model's responses to changes in matrix porosity, fracture porosity, fracture permeability and thickness were carried out for each aquifer for the operational cycles. In addition, sensitivity analyses to changes in matrix diffusivity and dispersivity were undertaken for the Chalk aquifer.

The results showed low sensitivity to parameters in the estimated ranges with only matrix porosity having a significant effect in the Chalk, the Lincolnshire Limestone and the Sherwood Sandstone.

The main conclusion is that dual-porosity aquifers will need more conditioning than single porosity aquifers. However, the suitability of any aquifer depends on the dispersive and diffusive parameters which can generally only be obtained from field trials.

SWIFT cannot incorporate geochemical reactions, both water/water and water/rock, which can be important in determining recovered water quality. PHREEQC is a reactive model which can quantify processes likely to occur in ASR schemes. In a non-reactive mode, PHREEQC was successfully calibrated to the output from SWIFT and sensitivity analysis on the physical aspects of the model was executed in four stages of increasing complexity.

PHREEQC has several limitations in describing the physical aspects of an ASR-cycle including:

- a simplified conceptualisation of dual porosity.
- inability to define injection or recovery rate.
- conceptualisation of radial flow in a one-dimensional model.

A variety of techniques were used to overcome these limitations including a thorough calibration to the results of the SWIFT model.

Linking the two models successfully should enable both the physical and geochemical aspects of ASR to be simulated. This will provide a valuable tool to assist site selection and design of testing and operational schemes. The programme of continued research and development is identified in order to achieve the objectives of the project.

## **1. INTRODUCTION**

The aim of this report is to explore and assess the possibilities of applying physical and geochemical models to Aquifer Storage and Recovery (ASR). Such models can be applied for testing purposes both during the development of the site as well as for operational purposes once the development is completed.

The report consists of three parts

- (i) The development of the modelling of physical aspects of ASR using SWIFT for a variety scenarios, based on generic data, for the Chalk, the Lincolnshire Limestone, the Sherwood Sandstone and the Upper Greensand. A preliminary assessment of the potential of these aquifers for ASR purposes is obtained.
- (ii) The development of a model with PHREEQC for conservative transport during ASR cycles. PHREEQC is capable of incorporating water quality aspects of ASR caused by chemical reactions (e.g. precipitation, dissolution, redox ...). However, this first step focuses only on physical aspects of transport.
- (iii) The calibration of the results for a conservative tracer of the 1-dimensional geochemical PHREEQC model (Parkhurst & Appelo, 1999) to the radial 2-dimensional SWIFT model.

Recommendations for the next steps in improving the models and their applicability are included.

## **2. PHYSICAL MODELLING**

### **2.1 The SWIFT Model**

SWIFT is a fully transient three-dimensional model which simulates the flow and transport of fluids (brines), heat (energy) and radionuclide chains in fractured geologic media. It is extremely appropriate for modelling ASR in fractured media because of its ability to model dual porosity effects in aquifers. These are likely to influence the recovered water during an ASR-scheme and are a widespread phenomenon in UK aquifers (Chalk, Lincolnshire Limestone). When a dual porosity medium is modelled in SWIFT, the modelled system is described in two parts: the fractures and the porous blocks. These are called “global” and “local” systems respectively. Both systems have their own properties which must be defined for the entire modelled region.

For an ASR-test site in the Chalk, the model was calibrated based on measurements of 8 ASR-test cycles, and predictions were made for concentrations of chloride during the recovery of the 9<sup>th</sup> cycle. The close fit between modelled and measured concentrations confirms the reliability of the model.

### **2.2 Modelling the Major British Aquifers**

In a first attempt to assess the physical impact of ASR-schemes on the recovered water quality in the British aquifers a sensitivity analysis was carried out with the SWIFT-model. Based on estimates of aquifer parameters the impact of operational and test cycles on the recovered water quality were modelled. The results (the concentrations measured in the well) show the possible range in recovered water quality within each aquifer and the differences in recovered water quality between different aquifers. No chemical reactions between the injected water and the native groundwater and the aquifer material are included in the model. The changes occurring reflect solely mixing processes.

The aquifers considered for this work are the Chalk, Lincolnshire Limestone, the Lower Greensand and the Sherwood Sandstone, these being the aquifers most likely to be used for ASR. The Chalk and the Lincolnshire Limestone are both best described as dual porosity aquifers. The main difference between both is the matrix porosity (2 to 40 times greater in the Chalk). Dual porosity means that the bulk of the flow takes place in the fractures and that storage is provided by the matrix. The important parameters in this type of aquifer, as far as solute transport is concerned, are the fracture and matrix porosity, the spacing between the fractures and the diffusion coefficient, which is a measure of the rate at which solute can penetrate the matrix blocks. Of these parameters only matrix porosity is readily measured, although estimates of fracture porosity and fracture spacing can be made from pumping test data and from geophysical logging. The diffusion coefficient of some solutes into clean chalk samples have been measured and so estimates (order of magnitude) for this parameter are available. However the relevance of laboratory measurements on clean uniform surfaces to processes occurring at natural fracture surfaces is poorly understood and is at present being investigated.

In contrast to the Chalk and Limestone, the Lower Greensand behaves as a single porosity medium. This means that the same void spaces (as measured by porosity) control both the permeability and the storage. In this case the parameters which are of most relevance to transport of solutes in the porosity and the dispersivity. Dispersivity is a measure of how much a plume of solute will spread when transported and is controlled by the heterogeneity of the aquifer at all scales. Dispersivity can be measured in the laboratory under controlled conditions but the values obtained are found not to be relevant for larger scales. This is because of the large scale heterogeneities in the aquifer material and the flow field which are present in ‘uncontrolled’ field conditions. Dispersivity of an aquifer is usually obtained as a model calibration parameter.

The behaviour of the Sherwood Sandstone lies somewhere between the dual-porosity and single-porosity conceptualisations and is probably best described as a dual-permeability medium. This is because the matrix is permeable enough to transmit water at the low rates which are relevant in regional flow systems, but probably not in the near-well situation, where fracture flow is likely to dominate. The SWIFT model cannot simulate this type of aquifer, and as such it may not be an appropriate model for simulating ASR behaviour in the Sherwood Sandstone. In the work reported here the Sherwood Sandstone has been modelled as a dual-porosity medium, but further work is planned to model this aquifer more accurately.

### 2.2.1 *Parameter estimates*

An analysis was carried out to estimate a range for the main parameters in the four major aquifers (Chalk, Lincolnshire Limestone, Sherwood Sandstone, Lower Greensand) based on data from (Allen et al., 1997). The estimated parameters or parameter ranges for the different aquifers are indicated in Table 1 and encompass:

- permeability
- porosity
- specific storage
- thickness

for porous aquifers, and

- matrix permeability
- fracture permeability

- matrix porosity
- fracture porosity
- specific storage
- thickness of the aquifer

for dual porosity aquifers.

**Table 1** Estimated values or ranges of values for the parameters of the main aquifers

Aquifer	Type	Permeability (m/d)		Porosity (%)		Storativity ( $m^{-1}$ )	Thickness (m)
		Matrix	Fracture	Matrix	Fracture		
Lincs Limestone	D	$10^{-3}$	10-100	1-10	0.5-5	$10^{-6}$	10-40
Chalk	D	$10^{-3}$	1-100	20-40	0.5-2	$10^{-6}$	50-200
Sherwood Sandstone	D	1-50	10-500	10-25	1	$10^{-5}$	50-300
Lower Greensand	S	1-100		10-25		$10^{-5}$	10-40

In addition to these parameters, the recovered water quality also depends on other physical parameters for which no estimates based on direct in situ measurements are possible. These are:

- the dispersivity
- the matrix diffusivity (dual porosity aquifers only)
- the fracture diffusivity (dual porosity aquifers only)
- the molecular diffusion (porous aquifers only)

These values are often estimated during model calibration.

### 2.2.2 Sensitivity analysis

Before considering the sensitivity of the model to the various parameters it is worthwhile examining the processes which occur during an injection-abstraction cycle in a dual porosity aquifer and the form of the output produced by the model (Figure 1). The system that is being modelled is a cylinder of fractured aquifer centred around a well. The aquifer starts off full of water of a salinity 1.0 – native groundwater. This water is in the fractures and the matrix. Water of salinity 0.0 is pumped into the aquifer through the well. As the fractures are far more permeable than the matrix, this water flows in the fractures away from the well thus reducing the salinity of the water in the fractures (Figure 1). This means that the water in the matrix and the water in the fractures near the well have a different chemical composition, which causes a diffusive exchange to occur between these two water bodies. This makes the water in the matrix slightly less saline and that in the fractures slightly more so. When injection stops and the system is allowed to stand (Figure 1), flow in the fractures also stops but the diffusive exchange continues, causing the fluid in the fractures which have been affected by the injection, to become increasingly contaminated by the fluid in the matrix. Meanwhile the matrix fluid becomes less contaminated. The rate at which this diffusive exchange occurs depends on the

diffusivity of the matrix. When recovery starts, water is drawn back along the fractures towards the well. This water will not be of the same chemistry as the injected water causing the salinity measured at the well to increase. Whilst pumping continues, water from further out in the aquifer will be drawn towards the well. This will come into contact with matrix water which has been diluted by its previous contact with the injection water. In this situation the contaminants in the fracture water will diffuse into the matrix, which will lower the salinity of the fracture water slightly. At the end of the abstraction period, the aquifer contains fracture water and matrix water which are not in chemical equilibrium and so is subject to further diffusive exchange.

For each aquifer a base case was modelled. The parameter values were chosen to be the middle of the estimated parameter range (if a range was indicated). Starting from this base case a sensitivity analysis was carried out for each parameter. Therefore, the model was run for several parameter values within the estimated range. The parameters values are shown in Table 2.

**Table 2 Summary of the modelled parameter values for the four aquifers**

	<b>Lincoln Limestone</b>	<b>Lower Greensand</b>	<b>Chalk</b>	<b>Sherwood Sandstone</b>
Matrix porosity (%)	1-5-10	10-15-20-25	20-25-30-35-40	10-15-20-25
Matrix permeability (m/d)	-	10-20-40-60-80-100	-	1-10-20-30-40-50
Fracture porosity (%)	0.5-1-2-3-4-5	-	0.5-1-1.5-2	
Fracture permeability (m/d)		-	1-20-40-50-60-80-100	10-50-100-150-200-250-300
Thickness (m)		10-25-40	50-100-150-200	50-100-200-300

For the sensitivity analysis, two ASR regimes were tested. One regime represents a typical series of test cycles (short cycles), while the second represents a typical series of operational cycles (long cycles) (Table 3). This was done to ascertain whether any of the parameters had larger effects on different timescales.

**Table 3 Summary of the ASR-schemes used for the sensitivity analysis**

	<b>Test Cycle</b>				<b>Operational Cycle</b>
	<b>In</b>	<b>Stand</b>	<b>Out</b>	<b>Stand</b>	5 cycles of 1 year each, including:
Cycle 1	5 days	Overnight	7 days	Overnight	<ul style="list-style-type: none"> <li>• injection: 4 months</li> <li>• stand: 3 months</li> <li>• recovery: 3 months</li> <li>• stand: 2 months</li> </ul>
Cycle 2	7 days	Overnight	5 days	Overnight	
Cycle 3	10 days	Overnight	5 days	Overnight	
Cycle 4	10 days	Overnight	5 days	Overnight	
Cycle 5	10 days	5 days	5 days	Overnight	
Cycle 6	10 days	5 days	10 days		

After a complete sensitivity analysis for the Chalk parameters was carried out, it became clear that the sensitivity of most parameters was very low. Therefore, during the analysis of the other aquifers, only the parameters with a range which showed little or no overlap with the parameter range for the Chalk were modelled and only the operational ASR-scheme was tested. It was assumed that if the sensitivity of a parameter was low during long cycles, it would be even lower during the test-cycles. The analysis which were finally carried out are indicated in Table 4.

**Table 4** Summary of the executed sensitivity analysis

	<b>Lincolnshire Limestone</b>	<b>Upper Greensand</b>	<b>Chalk</b>	<b>Sherwood Sandstone</b>
Operational Cycles	#	*	*	*
Test Cycles			*	
* all parameters included in the analysis				
# only influence of matrix porosity and fracture porosity evaluated				

In order to complete the analysis for the Chalk, the influence of physical parameters for which no empirical estimates are possible were evaluated. These are:

- matrix diffusivity within the range of  $10^{-8}$ - $10^{-10}$   $m^2s^{-1}$
- dispersivity within the range of 0.1–10 m

### 2.2.3 Interpretation and conclusion

The results are initially interpreted for each aquifer separately, followed by a comparison of the results for the different aquifers. The results are expressed as relative concentrations against time, where the concentration of the injected water is 0 and the concentration of the native groundwater is 1. The results are for a non-reactive solute only.

#### *Chalk*

In general, if the parameter values are chosen within the estimated range, then their sensitivity is low and the changes in relative concentration of a conservative parameter are small.

Because of these low sensitivities, the conclusions drawn for the short term test cycles can be extrapolated to long term operational cycles as illustrated in Figures 2 and 3.

From the estimated parameters only the matrix porosity (Figure 2) seems to have an influence on the modelled relative concentrations. The other parameters (fracture permeability, fracture porosity and thickness of the aquifer) cause only minor differences

Figure 4 indicates that the sensitivity to the parameters which are difficult to estimate (matrix diffusivity, dispersivity), if their estimated range is correct, can be higher than the sensitivities for the parameters shown in Table 1.

It cannot simply be concluded that the parameters in Table 1 do not influence the modelled concentrations because the dispersivity and matrix diffusivity depend on the values of the parameters in Table 1 (e.g. such as fracture permeability, rock porosity and matrix porosity).

### *Sherwood Sandstone – Lincolnshire Limestone (Figure 5 and 6)*

The same conclusions as for the Chalk can be drawn. Only matrix porosity influences the modelled concentrations significantly.

### *Lower Greensand (Figure 7)*

For the Lower Greensand none of the tested parameters have a significant influence on the modelled concentrations when chosen within the estimated ranges.

### *Comparison of the base cases for each aquifer*

The modelled base case for each aquifer is presented in Figure 8. Based on the parameter estimates the Chalk seems to be the least suitable for ASR-purposes in terms of recovered water quality based on physical processes. This is due to its high matrix porosity in comparison with the other aquifers leading to a high degree of mixing of injected and native water. Due to the low matrix porosity, for the Sherwood Sandstone and the Lincolnshire Limestone, matrix effects are less important. The total absence of dual porosity effects in the Lower Greensand results in the quality of the native groundwater having a minimal influence on the recovered water.

The lack of sensitivity to the aquifer thickness and the permeability is not unexpected in this modelling exercise, because the same volume of water has been injected in all the model runs. This, of course, leads to very different imposed heads on the aquifer, some of which would be impractical in reality. However the volume of the aquifer (and thus the volume of the native groundwater) which comes in contact with the injected water is the same in all the cases where only the permeability or the thickness is changed. For the dual-porosity aquifers, the matrix permeability has little effect on predicted concentrations because of the assumption in SWIFT that there is no flow in the matrix, except perpendicular to the fractures. This is a reasonable assumption for the Chalk and the Lincolnshire Limestone. Also the Sherwood Sandstone was modelled as a dual porosity aquifer, although this aquifer can better be described as a dual-permeability medium. However, SWIFT is not directly capable of modelling dual-permeability media.

The difference between the single and dual-porosity response of the aquifers is significant and so it is important to be certain that the conceptual model used for predictions is appropriate to the aquifer being tested.

The main conclusion of this modelling work is that dual-porosity aquifers will need more conditioning (injection of non recoverable water to build up a buffer zone of mixed waters) than single porosity aquifers before they are suitable for use for ASR. However the suitability of any aquifer depends on the dispersive and diffusive parameters which can generally only be obtained from field trials.

### *Further steps to be undertaken*

The influence of matrix diffusivity and dispersivity was evaluated only in the case of the Chalk. Both parameters are expected to depend on the parameters values taken into account for the sensitivity analysis. However, for all simulations matrix diffusivity and dispersivity were assumed to be constant because their dependency is unknown and can therefore not be incorporated. The relationship between matrix diffusivity and dispersivity and the parameters in Table 1 should be investigated to obtain better estimates for the first parameters.

The Sherwood Sandstone aquifer was modelled as a dual porosity aquifer although this aquifer can be seen as an intermediate between a dual porosity aquifer and a single porosity aquifer (dual permeability medium). Further modelling for the Sherwood Sandstone is necessary to obtain a better

conceptualisation for this aquifer where flow is expected to take place both in the fractures and in the matrix.

### **3. PHYSICAL MODELLING WITH PHREEQC**

Several ASR-field trials have indicated that physical modelling alone cannot always predict the quality of recovered water. This is because chemical reactions cannot be incorporated in these models. Depending on the chemistry and mineralogy of the sediment and the quality of the native and the injected water a wide range of chemical reactions can influence the quality of the recovered water. To model these reactions a geochemical transport model is necessary. However, although geochemical models have a large scope for modelling chemical interactions between sediments and water and between different types of water including both equilibrium and kinetic aspects, their tools to model more complex transport regimes are limited. Therefore, they cannot replace the flow-transport models and it should be investigated accurately if the modelled concentrations in the geochemical model for conservative ions (such as chloride), which are expected not to take part in any of the chemical reactions match with the results of the transport model.

This section represents the first step in applying geochemical models for ASR purposes and includes only the modelling of the physical framework. Once the physical framework is established and found to be reliable, geochemical processes can then be modelled. This second step has yet to be investigated and is not included in this report.

#### **3.1 The PHREEQC Model**

PHREEQC is based on an ion-association aqueous model and has capabilities for (1) speciation and saturation-index calculations; (2) reaction-path and 1D-transport calculations involving reversible reactions, which include aqueous, mineral, gas, solid-solution, surface-complexation, and ion-exchange equilibria, and irreversible reactions, which include specified mole transfers of reactants, kinetically controlled reactions, mixing of solutions and temperature changes and (3) inverse modelling, which finds sets of mineral and gas mole transfers that account for differences in composition between waters, within specified compositional changes (Parkhurst and Appelo, 1999).

#### **3.2 ASR Cycle Modelling with PHREEQC**

The main questions which arise when modelling ASR-cycles with PHREEQC are:

- what is the uncertainty incorporated in the physical system (due to the transformation of a 2-dimensional radial problem to a 1 dimensional model and the different approach in solving the transport equations in different models)
- how is this uncertainty going to influence the modelled chemical processes
- in case of high uncertainty should we work with scenarios (worst physical case scenario/best physical case scenario)

The setting up of the physical part of the PHREEQC model was executed in four stages. Subsequent stages increase in complexity and in each stage the influence of newly introduced parameters on the modelled results were tested.

- (i) ASR-cycle (injection–stand-recovery-stand) without dual porosity and without radial flow (Stage 1)
  - check sensitivity of molecular diffusion and dispersivity on the modelled results

- (ii) ASR-cycle (injection–stand–recovery–stand) including dual porosity but without radial flow (Stage 2)
  - check sensitivity of dual porosity parameters (mobile phase porosity, immobile phase porosity, exchange factor)
  - check the influence of the number of cells
- (iii) ASR-cycle (injection–stand–recovery–stand) without dual porosity but including radial flow (Stage 3)
  - set up Excel file to generate cell lengths
  - check the influence of radial/normal cells.
- (iv) ASR-cycle (injection–stand–recovery–stand) including dual porosity and radial flow - Extension to multiple ASR-cycles (Stage 4)

3.2.1 ASR-cycle (injection–stand-recovery-stand) without dual porosity and without radial flow (Stage 1)

ASR-cycle: injection for 50 days, stand for 25 days, recovery for 50 days, stand for 25 days.

Processes included: advection, dispersion and diffusion

**Table 5 PHREEQC parameters for Stage 1**

Parameter	Value	Parameter	Value
Number of cells	100	Injected Cl concentration	0 mg/l
Number of shifts*	102	Native Cl concentration	10 mg/l
Time step	86400 s (1 day)	Dispersivity	1 m
Length of the cells	2 m	Diffusion	$10^{-7} \text{ m}^2\text{s}^{-1}$
Total length	200 m		
Boundary conditions	constant flux	*This indicates the number of transfers from cell to cell	
Total porosity	0.3		

Sensitivities (Figure 9)

- Dispersivity influences both the shape of the curve and the maximum concentration during recovery. Higher dispersivities lead to a lower concentration during recovery, an earlier breakthrough of the native water during recovery and higher concentrations in the first cell during injection.
- When a reasonable value for the diffusion is defined ( $10^{-7} \text{ m}^2\text{s}^{-1}$ ) only very small retardation of the breakthrough curve is observed. The maximum concentration during the early recovery is lower but increasing slowly in later stages.

3.2.2 ASR-cycle (injection –stand –recovery –stand) including dual porosity but without radial flow (Stage 2)

ASR-cycle: injection for 50 days, stand for 25 days, recovery for 50 days, stand for 25 days.

Processes included: advection, dispersion and diffusion. Dual porosity is included with one matrix cell connected to each fracture cell. The process is defined by the porosity of both the mobile and the immobile phase and an exchange factor.

**Table 6 PHREEQC parameters for Stage 2**

Parameter	Value	Parameter	Value
Number of cells	100	Injected Cl concentration	0 mg/l
Number of shifts*	102	Native Cl concentration	10 mg/l
Time step	86400 s (1 day)	Dispersivity	1 m
Length of the cells	2 m	Diffusion	$10^{-7} \text{ m}^2 \text{ s}^{-1}$
Total length	200 m	Porosity mobile phase	0.01
Boundary conditions	constant flux	Porosity immobile phase	0.3
Total porosity	0.3	Exchange factor	$10^{-9} \text{ s}^{-1}$

\* This indicates the number of transfers from cell to cell

Within the PHREEQC-model, dual porosity can be modelled in two ways. For this first approach, the first order exchange approximation is used. Using this approach the dual porosity effects are determined by an exchange factor ( $\alpha$ ) which depends on the diffusivity ( $D_e$ ), the porosity of the immobile phase ( $\theta_{im}$ ), a scale factor ( $a$ ) and a shape factor ( $f_{s \rightarrow 1}$ ) related to the “shape” of the matrix phase (see Parkhurst and Appelo (1999) p. 55). How this scale factor for the matrix relates to the definition of the matrix in SWIFT is not clear.

An estimate for the value of the exchange factor ( $\alpha$ ) can then be made based on the expression:

$$\alpha = (D_e * \theta_{im}) / (a * f_{s \rightarrow 1})$$

The diffusivity is expected to range between  $10^{-8} - 10^{-10} \text{ m}^2 \text{ s}^{-1}$ , the porosity of the immobile phase between 0.1- 0.4 and the scale factor ( $a$ ) being half of the cell length: 1 m.

In the case when defining the matrix as a planar sheet  $f_{s \rightarrow 1} = 0.53$ , and when defining the matrix as a rectangular (infinite) prism  $f_{s \rightarrow 1} = 0.312$ .

This corresponds to a smallest value of  $\alpha$  of  $2.8 \cdot 10^{-12} \text{ s}^{-1}$ , while the largest value is  $1.423 \cdot 10^{-8} \text{ s}^{-1}$ . For the calculations at this stage a value of  $10^{-9} \text{ s}^{-1}$  was used.

Sensitivities for the porosity of the mobile phase, the immobile phase, the exchange factor and the number of cells are indicated in Figure 10.

The exchange factor has an important influence on the modelled concentrations (Figure 10a). This can be summarised as:

*Low exchange factor:* the effect of dual porosity is minimised and the shape of the curve is almost the same as if no dual porosity medium is present.

*Medium exchange factor:* during the injection phase an exchange with the matrix takes place leading finally to a constant concentration; during the standing phase the influence of the matrix leads to quickly increasing concentrations, during the recovery the concentration increases further while in the following standing phase, exchange with the partly freshened matrix induces slightly decreasing concentrations.

*High exchange factor:* during the injection phase there is a intense exchange with the dual porosity cells leading to high concentrations, therefore in the subsequent standing phase no further exchange can take place. The same effect is observed during the recovery phase and the subsequent standing phase.

The higher the porosity of the mobile phase the less the influence of the matrix (Figure 10b). Low values lead to a high concentration during injection and a quick rebound while standing both after the injection and the recovery. High values lead to low chloride concentrations throughout the cycle and only a minor rebound during the standing phase after injection.

The value of the porosity of the immobile phase (Figure 10c) does not seem to have a direct effect on the modelled concentrations. However, the porosity of the immobile phase is a parameter necessary to calculate the exchange factor. Changes in the porosity of the immobile phase lead therefore to changes in the exchange factor and indirectly to changes in the modelled concentrations.

The influence of the number of cells/time step length (Figure 10d) is only visible during the injection phase; a lower amount of larger cells leads to slightly higher concentrations.

### 3.2.3 ASR-cycle (injection – stand –recovery –stand) without dual porosity but including radial flow (Stage 3)

ASR-cycle: injection for 50 days, stand for 25 days, recovery for 50 days, stand for 25 days.

Modelled Processes: advection, dispersion and diffusion ; radial flow

**Table 7 PHREEQC parameters for Stage 3**

Parameter	Value	Parameter	Value
Number of cells	100	Injected Cl concentration	0 mg/l
Number of shifts*	150	Native Cl concentration	10 mg/l
Time step	86400 s (1 day)	Dispersivity	1 m
Length of the cells	decreasing radially	Diffusion	$10^{-7} \text{ m}^2\text{s}^{-1}$
Total length	200 m		
Boundary conditions	constant flux		
Total porosity	0.3	* This indicates the number of transfers from cell to cell	

In SWIFT, the aquifer is modelled based on a two-dimensional radial flow model. Since PHREEQC is a one-dimensional model the same approach is not possible. To simulate radial flow in PHREEQC subsequent cell lengths are chosen so that the according subsequent rings have a constant volume. This can be arranged by giving successive cells lengths based on the following expression (Appelo & Postma, p 437):

$$\text{Length (n)} = \text{Length (1)} \cdot (\sqrt{n} - \sqrt{(n-1)})$$

where n is the cell number.

A calculation spreadsheet was made wherein cell lengths are calculated when the length of the first cell and the amount of cells are defined.

The type of grid cells has an influence on the modelled concentrations when a high dispersivity is assumed (Figure 11). Comparison of both modelling results is ambiguous because the length of the first cell determines the injection/recovery rate (see 3.3.2) and cell lengths are different in both simulations. Differences in modelled concentrations might therefore also be a consequence of differences in rate. During the beginning of the injection phase concentrations are significantly lower for the radial grid in comparison to the normal grid due to the large cell lengths of the first cells. However, during the recovery and the subsequent standing period, concentrations are slightly higher. These differences are likely to diminish in the case where a smaller dispersivity is defined.

#### 3.2.4 ASR-cycle (injection–stand–recovery–stand) including dual porosity and radial flow - Extension to multiple ASR-cycles (Stage 4)

ASR-cycle: injection for 50 days, stand for 25 days, recovery for 50 days, stand for 25 days.

Processes included: advection, dispersion and diffusion; dual porosity is included with one dual porosity cell connected to each cell. (the process is defined by porosities of both the mobile and the immobile phase and an exchange factor); radial flow.

**Table 8 PHREEQC parameters for Stage 4**

Parameter	Value	Parameter	Value
Number of cells	100	Injected Cl concentration	0 mg/l
Number of shifts*	150	Native Cl concentration	10 mg/l
Time step	86400 s (1 day)	Dispersivity	1 m
Length of the cells	decreasing radially	Diffusion	$10^{-7} \text{ m}^2\text{s}^{-1}$
Total length	200 m	Porosity mobile phase	0.01
Boundary conditions	constant flux	Porosity immobile phase	0.3
Total porosity	0.3	Exchange factor	$10^{-9} \text{ s}^{-1}$

\* This indicates the number of transfers from cell to cell

The model was extended to 4 ASR-cycles (Figure 12a). In subsequent ASR cycles the influence of the matrix on the recovered water decreases slowly. The higher the ratio between injected and recovered amount of water, the lower the influence of the native groundwater during the recovery in

subsequent cycles. However, comparison of Figure 12a and b where the ratio is increased from 1 to 1.6 indicates that this injection of more water which cannot be recovered only leads to a marginal decrease of concentration after 4 cycles.

### 3.3 Calibration of PHREEQC to SWIFT Model Results

Since the PHREEQC model has several limitations in describing the physical part of an ASR-cycle a thorough calibration of the PHREEQC results to the results of the SWIFT model is necessary. This is because the SWIFT-model is assumed to be more accurate for modelling physical problems.

Possible shortcomings of the PHREEQC model might be:

- the conceptualisation of dual porosity in PHREEQC is simplified in comparison to the one in SWIFT
- within PHREEQC it is not possible to define an injection or recovery rate directly; determining the flow rate by the shifting of pore volumes from cell to cell induces difficulties
- the conceptualisation of radial flow in a one dimensional model

Calibration to assess the influence of these shortcomings can be done based on real data or on results from other models. In this case the calibration based on the results from the SWIFT-model from a realistic scenario is preferred to the calibration against real data. The main reason for that is that real data are scarce, incomplete and not ideal. Observations covering the whole curve (certainly also during the standing phases) of an ASR-cycle are necessary to assess the reliability of the results.

Therefore, an operational ASR scheme was devised and modelled with both models. All parameters were defined in such a way that exactly the same problem was modelled. The results of the PHREEQC model were then fitted to the results of the SWIFT model by adjusting a fitting parameter (chosen as the exchange factor) in the PHREEQC model.

#### 3.3.1 Description of the modelled problem and parameters

The parameters for the calibration problem are described in Table 9. Two ASR-cycles are modelled, each lasting for 1 year. A standing period is included before and after the recovery phase. The injection rate during the 4 months of injection is 450 m<sup>3</sup>/d for the whole thickness of the aquifer or 45 m<sup>3</sup>/d per fracture.

**Table 9 Specifications for the calibration problem**

SWIFT	PHREEQC
<b>Discretisation</b>	
2-D radial flow	1-D flow
Total distance: 2000 m	Total distance:
Number of cells: 100	Number of cells: 113
Number of dual porosity cells: 10	Number of dual porosity cells: 1
Length: logarithmic increasing:	Equal volume cells in the case of radial flow: first cell: 40.03 m
Boundary conditions: constant concentration	Boundary conditions: constant concentration
Thickness: 10 m	Thickness: none – 1D
Time stepping: chosen by the model	Time shifts: 96664 s
<b>Injection scheme</b>	
4 months injection – 2.6 months stand – 2.6 recovery- 2.6 stand (1 year complete cycle)	4 months injection – 2.6 months stand – 2.6 recovery- 2.6 stand (1 year complete cycle)
2 cycles (2 years)	2 cycles (2 years)
<b>Rock properties</b>	
Compressibility: $4.4 \cdot 10^{-10} \text{ Pa}^{-1}$	-
Density: $1000 \text{ kg m}^{-3}$	-
<b>Global System (fractures) properties</b>	
permeability: $6 \cdot 10^{-5} \text{ ms}^{-1}$	-
fracture porosity: 0.01	fracture porosity: 0.01
long/trans dispersivity: 5 m	dispersivity: 5 m
diffusivity: $10^{-9} \text{ m}^2\text{s}^{-1}$	diffusivity: $10^{-9} \text{ m}^2\text{s}^{-1}$
<b>Local System (matrix) properties</b>	
Compressibility: $10^{-9} \text{ Pa}^{-1}$	-
Matrix porosity: 0.2	Matrix porosity: 0.2
Block size: 1 m	
-	Exchange factor: calibrated value: $5e^{-8} \text{ s}^{-1}$
Matrix permeability: $10^{-8} \text{ ms}^{-1}$	-
Matrix diffusivity: $10^{-9} \text{ m}^2\text{s}^{-1}$	-
<b>Injected amount</b>	
450 m <sup>3</sup> /d (1 fracture/m, 10 m aquifer thickness)	45 m <sup>3</sup> /d (1 fracture)

### 3.3.2 Calculation of the time shift in PHREEQC to make the flow rate in accordance with the defined injection/recovery rate in SWIFT

In order to make the injected amount in the PHREEQC model (where no direct injection/recovery rate can be defined) to agree with the injection rate from the SWIFT model, time shifts and number of time shifts have to be calculated appropriately. The injection rate in PHREEQC can only be introduced by defining the rate by which the pore volumes are moved from one cell to another. Therefore the pore volume of the first cell is calculated and the time step is chosen according to the injection rate wanted.

Consequently changing the injection/recovery rates or the fracture porosity during the modelling process requires a recalculation of the time stepping. It also means that it is not possible to define large differences in injection rates or large differences in injection and recovery rates in one run, because the maximum number of cells is limited. This limits the possibilities for modelling real cases considerably.

**Table 10** Calculation of time shifts and time stepping in PHREEQC according to a defined injection-recovery rate in SWIFT of 45 m<sup>3</sup>/d

Select the length of the first cell	40.0	m
Bulk volume of the first cell	5034.6	m <sup>3</sup>
Fracture porosity	0.01	
Pore volume of the first cell	50.346	m <sup>3</sup>
Injection rate (m <sup>3</sup> /d)	45	m <sup>3</sup> /d
Length of the time shift (d)	1.12	d
Length of the time shift (s)	96664.308	s
Length of the longest simulation	10800658	s
Number of time steps	112	
Number of cells required	113	

### 3.3.3 Modelling dual porosity in PHREEQC –calibration of the exchange factor

The way dual porosity is incorporated in the two models is slightly different. In SWIFT the local (matrix) system is modelled by attaching a one-dimensional grid to each of the global system (fracture) cells. The exchange between the cells of the matrix is defined by the matrix diffusivity. In PHREEQC dual porosity is modelled using a first order-exchange approximation as a mixing process between a mobile and a stagnant cell. For each cell in the model there is one corresponding stagnant cell. As well as this method, PHREEQC is also capable of modelling dual porosity based on the superposition of a finite difference grid on the stagnant zone. However, defining this finite difference grid is time-consuming and this method requires considerably more calculation time.

In this case the easiest approach, first order exchange approximation, was tested. The exchange factor was used to calibrate the PHREEQC to the SWIFT results.

It was illustrated earlier (Figure 10a) that the sensitivity of the exchange factor is high. To fit the PHREEQC results accurately to the SWIFT results a detailed sensitivity analysis was carried out in the range  $10^{-5} \text{ s}^{-1} - 10^{-11} \text{ s}^{-1}$ . The exchange factor influences the shape in a discontinuous

manner(Figure 13a). A value of  $5.10^{-8} \text{ s}^{-1}$  leads to the optimal fit although the first part of the curve (during injection) clearly deviates from the SWIFT-curve (Figure 13b).

#### 3.3.4 *Adjusting the time stepping*

The fact that the ratio of subsequent cell lengths must be related to the ratio of subsequent rings around a well to maintain the condition of radial flow, leads to large cells close to the well. This might induce the following problems:

- numerical dispersion related to the dispersivity
- calculations are carried out for the middle of each cell, the larger the cell the farther away from the well the concentration is calculated

In several model trials it became clear that reducing the dispersivity from 5 m to 0.1 m did not influence the results considerably. Therefore, it seems likely that the large volume of the first cells leads to an important influence of the matrix on the cells because these cells represent a large area around the well and a steep gradient of increasing matrix influence is smeared out over this large area.

It was attempted to reduce the size of the first cell and the subsequent cells. However, reducing the length of the cells needs also a recalculation of the time shift length and therefore an increased amount of cells necessary to allow for more time steps. PHREEQC seems to have a limit on the length of the cells (for this problem) that once the cells are smaller than 1 m a large amount of calculations thereby increasing the calculation time to several hours. Therefore, a compromise has to be sought between length of the first cell and acceptable calculation time. The results for three model runs are shown (Figure 14): an initial length of 50 m leads to a simulation time of less than 2 minutes, a length of 40 m requires 12 minutes and a length of about 31 m requires approximately 50 minutes. The results show that the reduced cell lengths lead to a better fit of the first and second stage of the curve with the PHREEQC results with an almost identical fit for the 3<sup>rd</sup> and 4<sup>th</sup> part of the curve. Further improvement is not possible because of these calculation time constraints.

#### 3.3.5 *Interpretation of the 1 cycle result- 2 cycle result*

A maximum deviation between the SWIFT and PHREEQC result can be seen in the beginning of the cycle where there is +/- 20 % difference (Figure 15a). During the rest of the modelled cycles differences are mainly less than 5 %, except for the standing phase after the recovery where SWIFT concentrations are a maximum of 10% higher than PHREEQC-concentrations. The overestimation of the concentration in PHREEQC during the injection and first standing phase and the underestimation during the subsequent phases point to the fact that the influence of the matrix on the result in PHREEQC is still too large in comparison to the SWIFT result. The same trend can be observed for the two-cycle model although the fit during the beginning of the second cycle is better than for the first cycle (injection and standing phase). This is caused by the reduced concentration in the matrix during the second cycle.

#### 3.3.6 *Improvement of the comparison of both models*

Since the concentration in the well itself cannot be modelled with the PHREEQC-model because of the large cell lengths, an optimised comparison between the two models was achieved by introducing a piezometer in the SWIFT-model (Figure 15b). The distance of the piezometer from the well is the same as half the cell length of the first cell in the PHREEQC-model (in this case 20 m). By comparing the modelled concentrations in the piezometer a better fit between SWIFT and PHREEQC was obtained (although the concentration in PHREEQC is still an average one over 40 m (being the length of the first cell)). The maximum concentration differences are now less than 5 %. The remaining differences can be interpreted as a consequence of the way the dual porosity is modelled in

both models. The presence of a gradient of mixing in the matrix in the SWIFT model buffers the effect of the matrix on the water quality in the fractures. The direct mixing in PHREEQC emphasises the matrix influence leading to more rapid concentration changes when the flow regime changes.

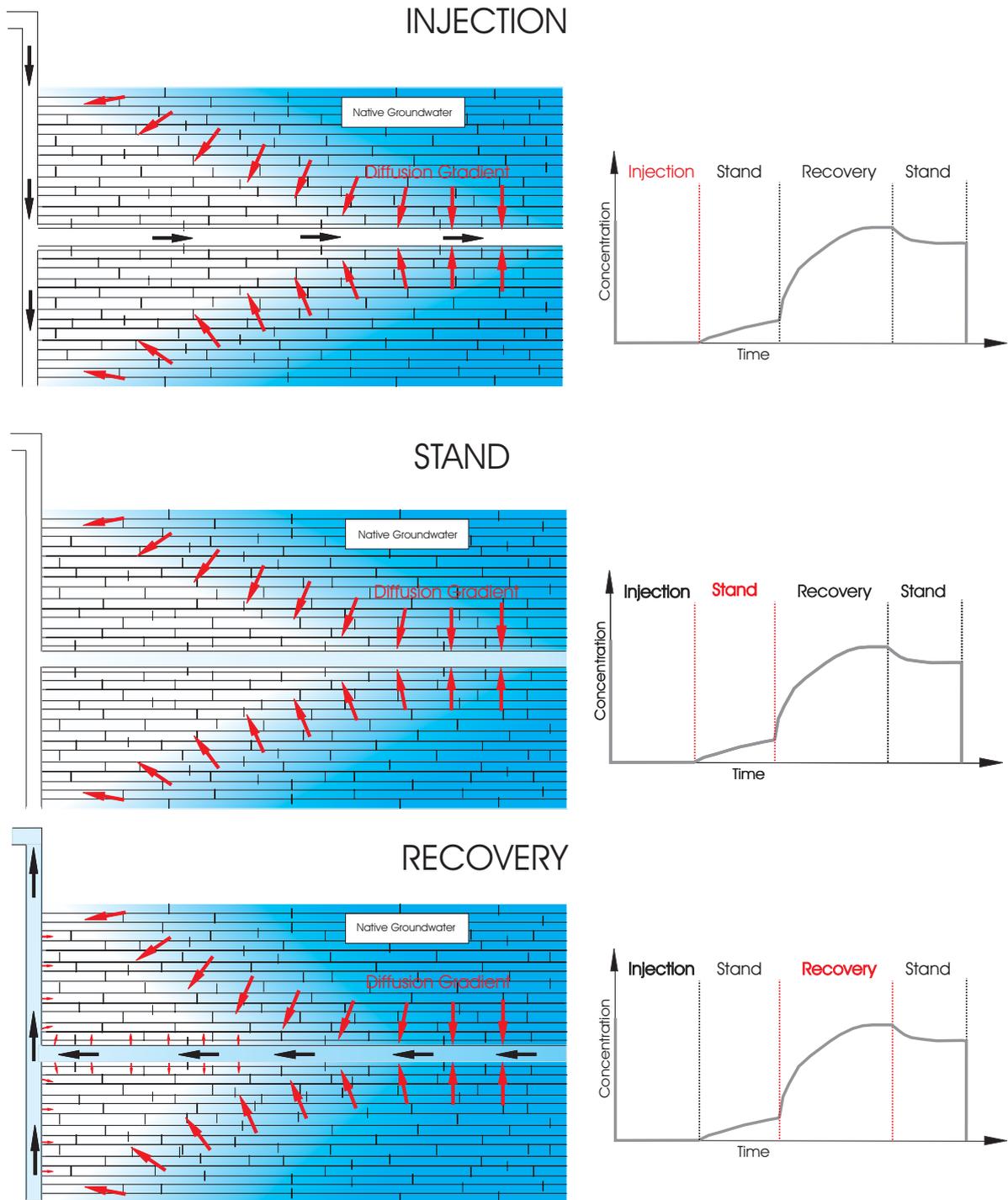
### **3.4 Further Development of the Geochemical Model**

In a next stage of the research programme the following topics should be investigated:

- Further calibration of the models with emphasis on:
  - influence of changing parameter values in both models
  - influence of longer or more ASR-cycles
- Is there a possibility to calibrate the model with a real dataset? (taking into account the limits for injection and withdrawal rates)
- What are the geochemical reactions to first build into the model e.g. mineral dissolution/precipitation, ion exchange, sorption, redox?

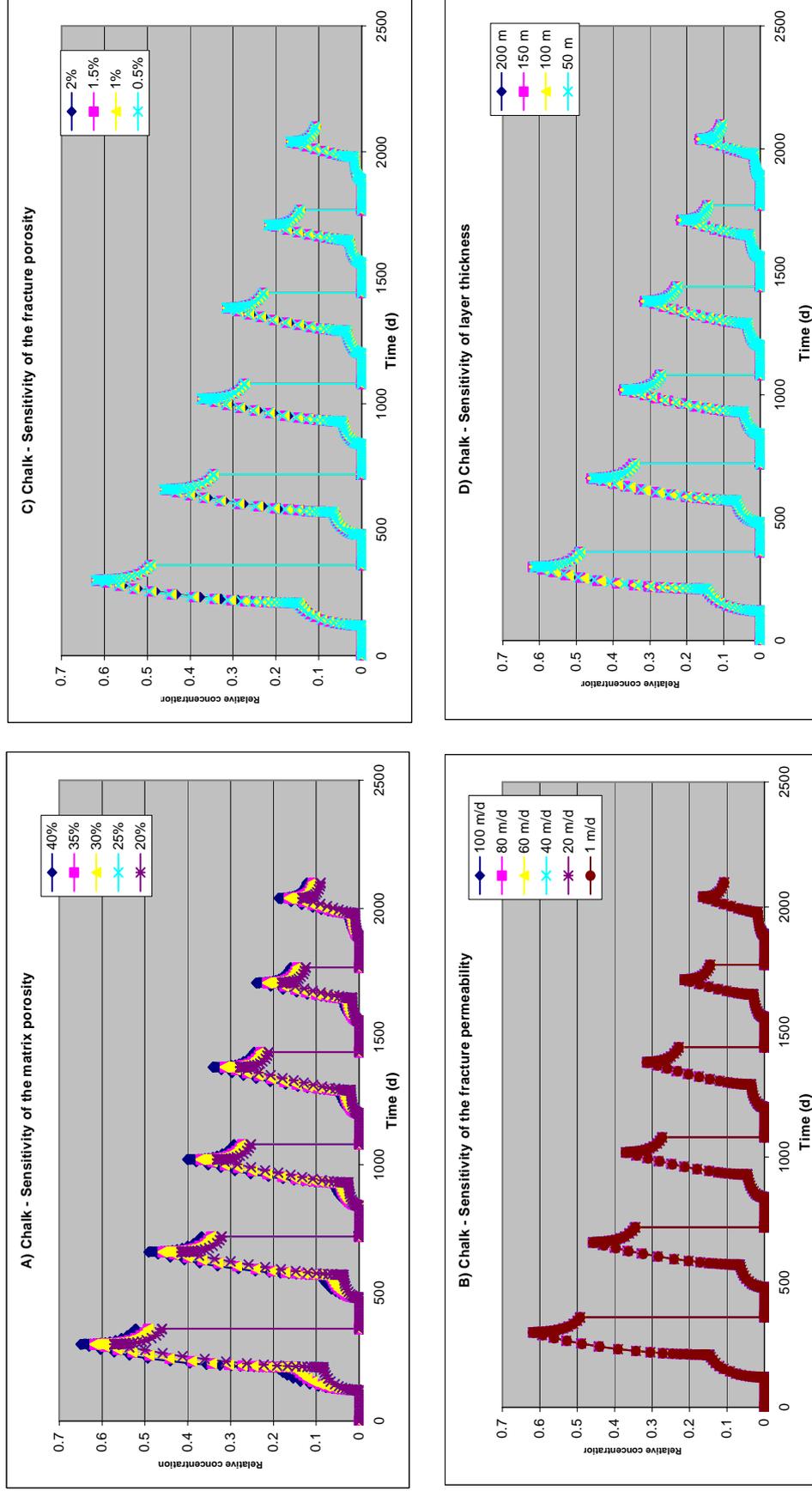
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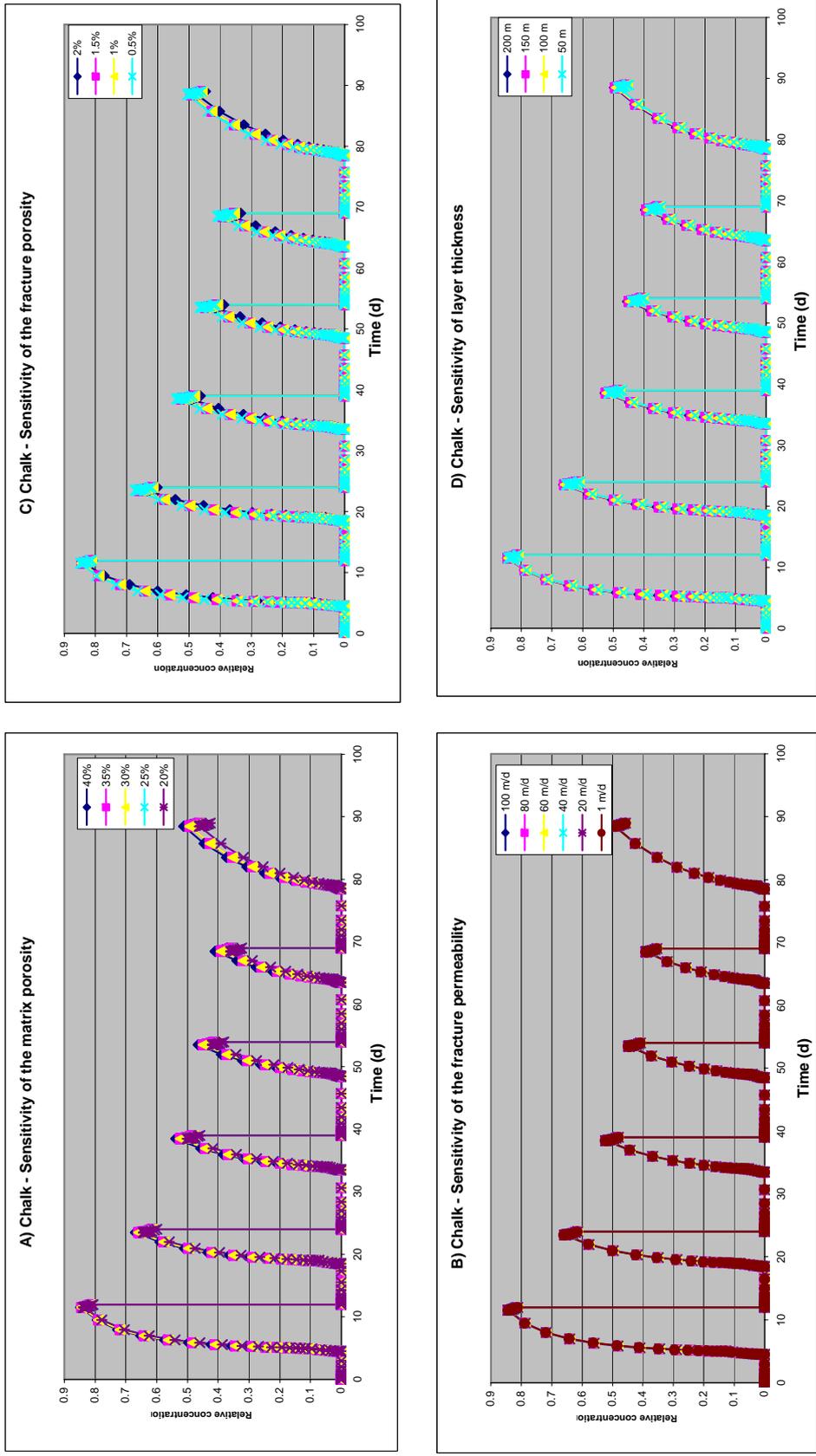
**Figure 1** Dual porosity effects illustrated for each stage of an ASR cycle

## OPERATIONAL CYCLES



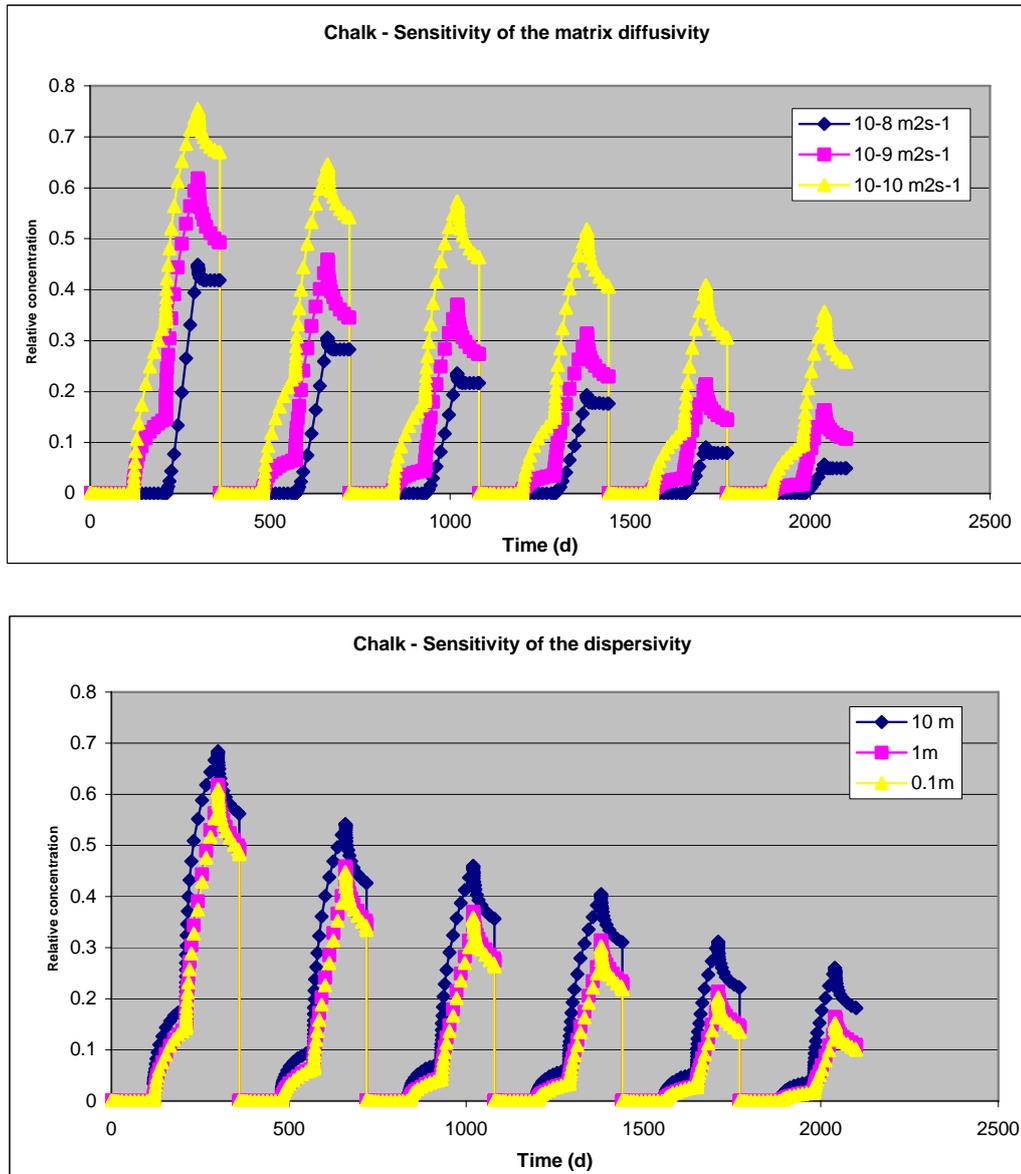
**Figure 2** Sensitivity of the matrix porosity, fracture permeability, fracture porosity and layer thickness during ASR-operational cycles in the Chalk

## TEST CYCLES



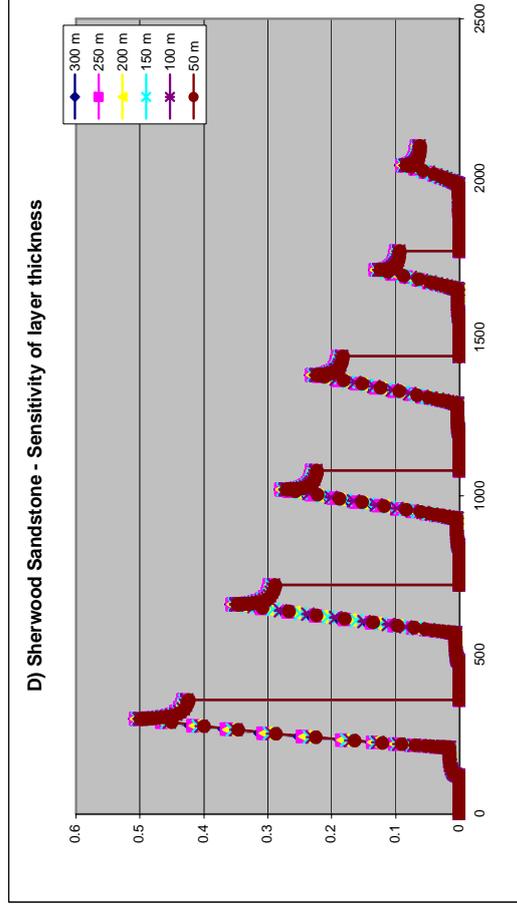
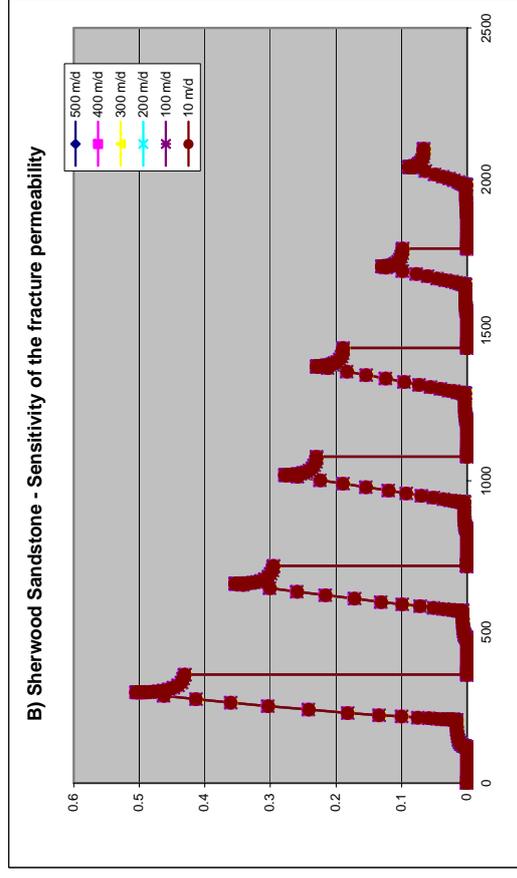
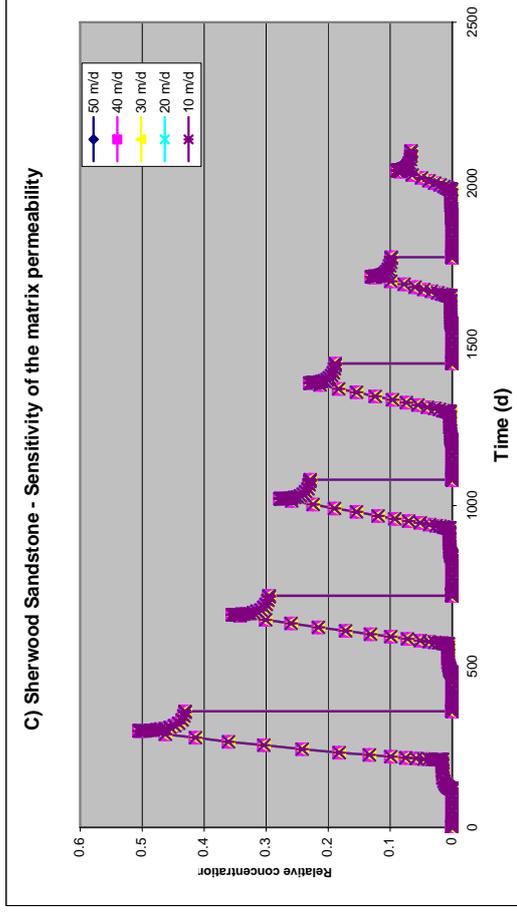
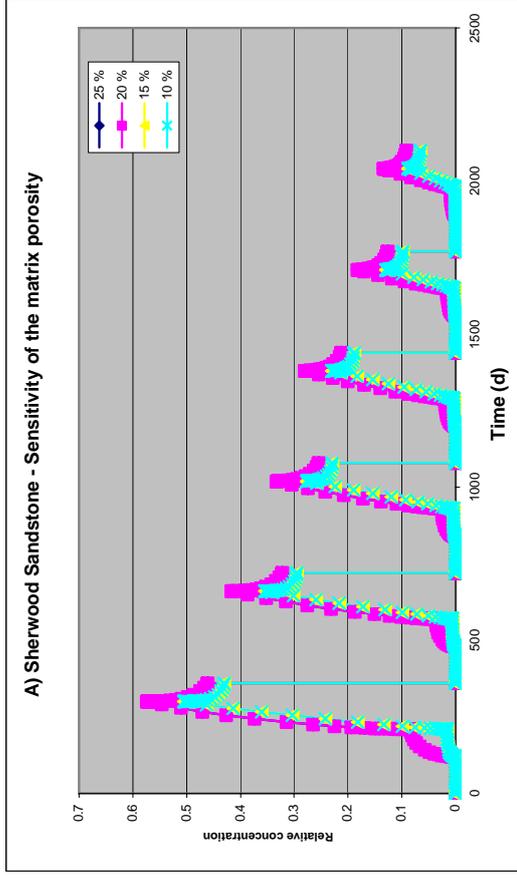
**Figure 3** Sensitivity of the matrix porosity, fracture permeability, fracture porosity and layer thickness during ASR-testing cycles in the Chalk

## OPERATIONAL CYCLES



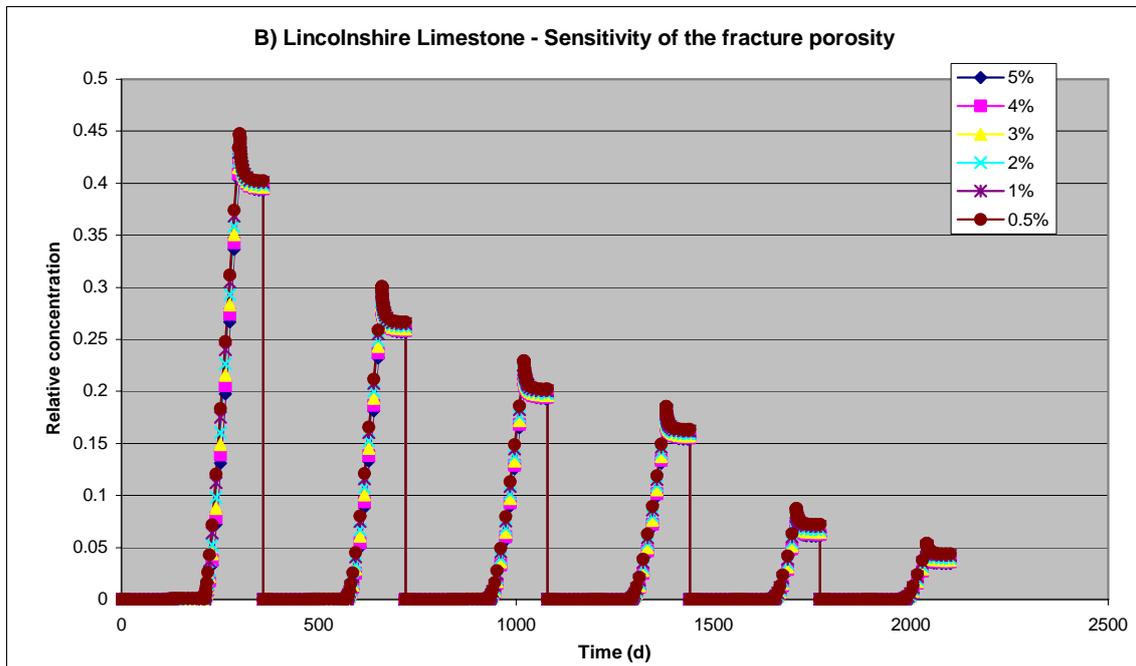
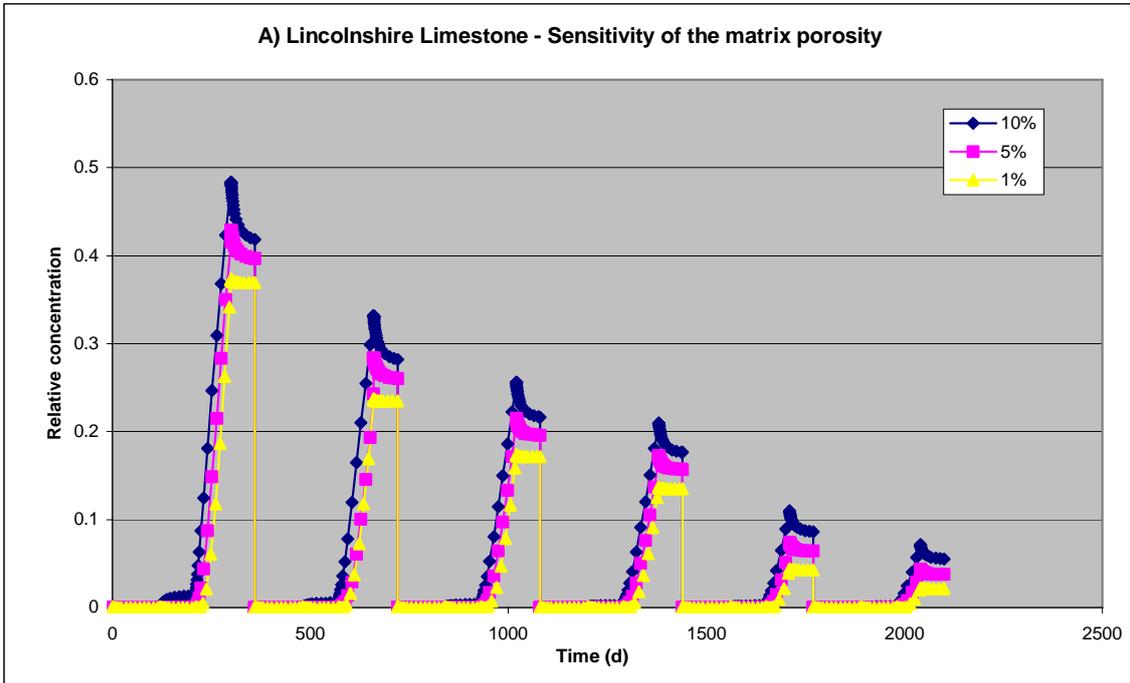
**Figure 4** Sensitivity of the matrix diffusivity and dispersivity during ASR-operational cycles in the Chalk

## OPERATIONAL CYCLES



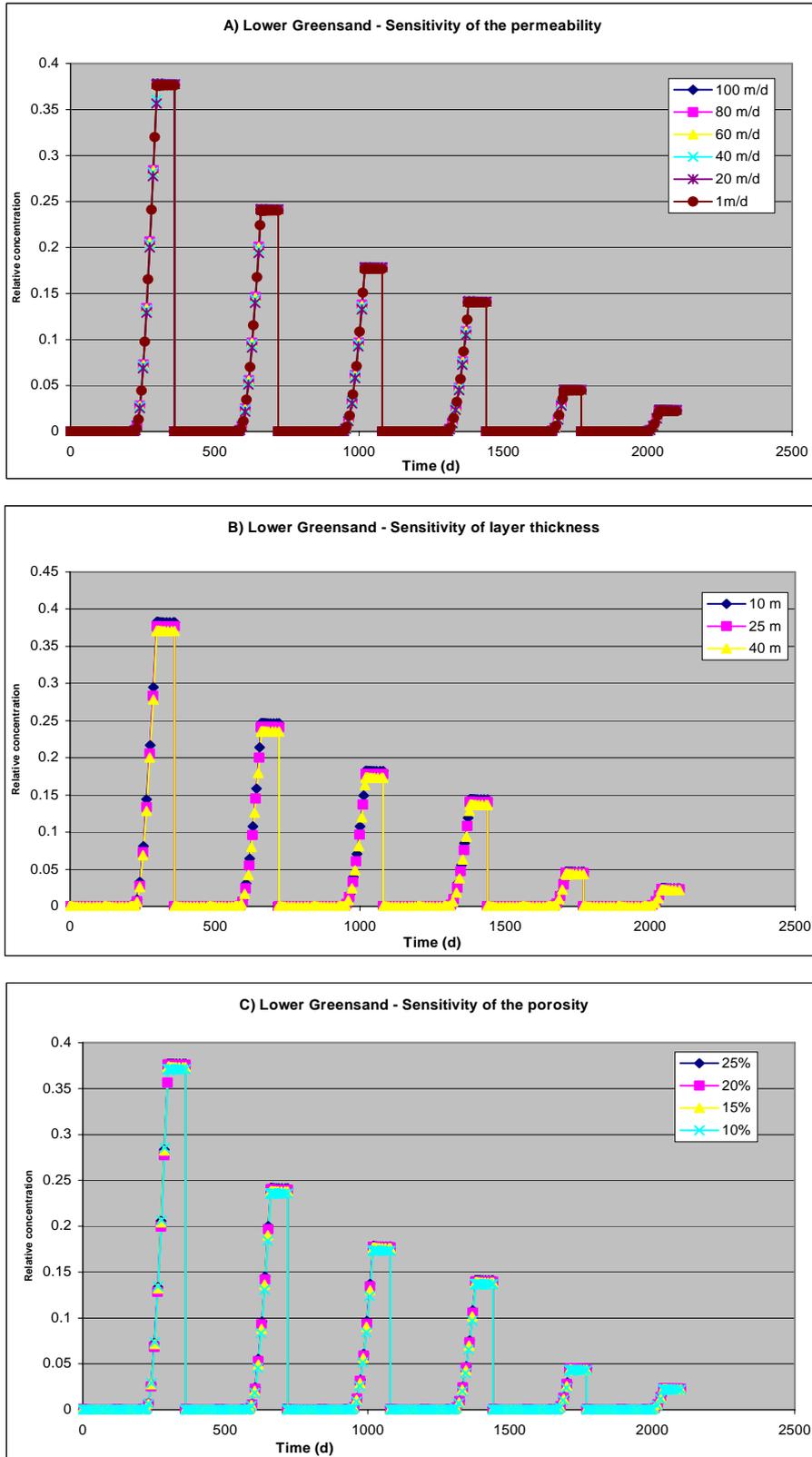
**Figure 5** Sensitivity of the matrix porosity, fracture permeability, matrix permeability and layer thickness during ASR-operational cycles in the Sherwood Sandstone

### OPERATIONAL CYCLES

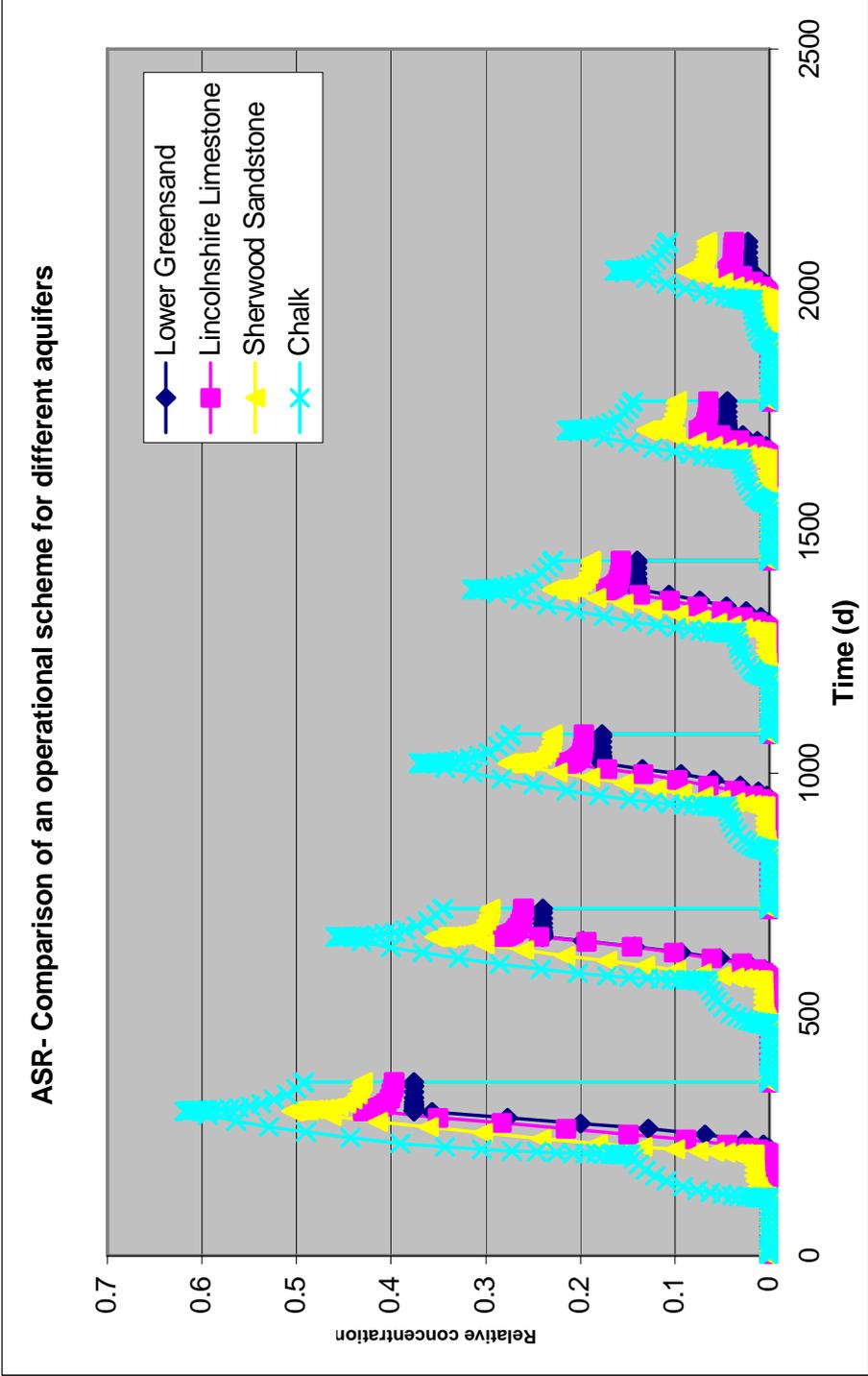


**Figure 6** Sensitivity of the matrix porosity and fracture porosity during ASR-operational cycles in the Lincolnshire Limestone

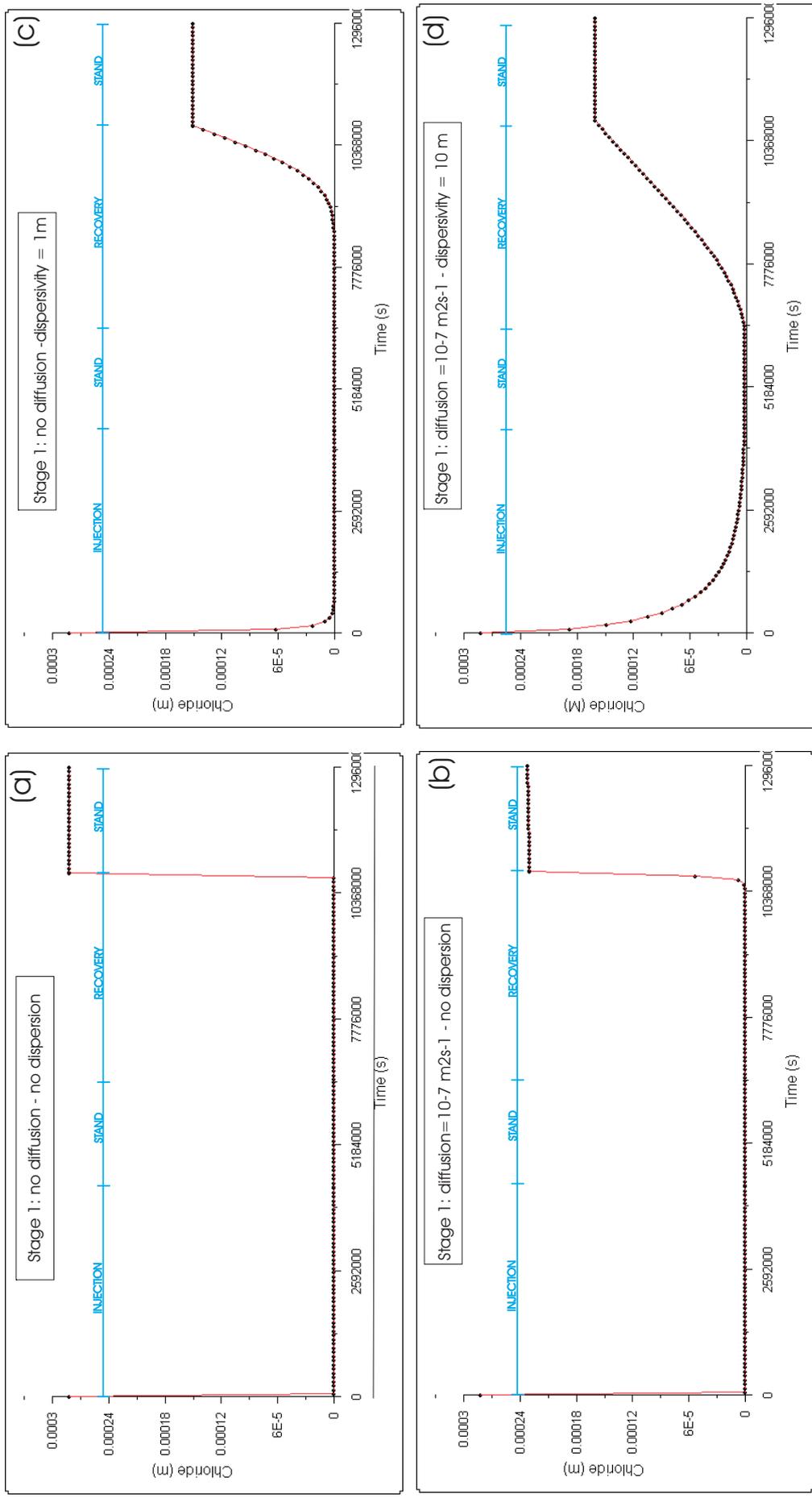
## OPERATIONAL CYCLES



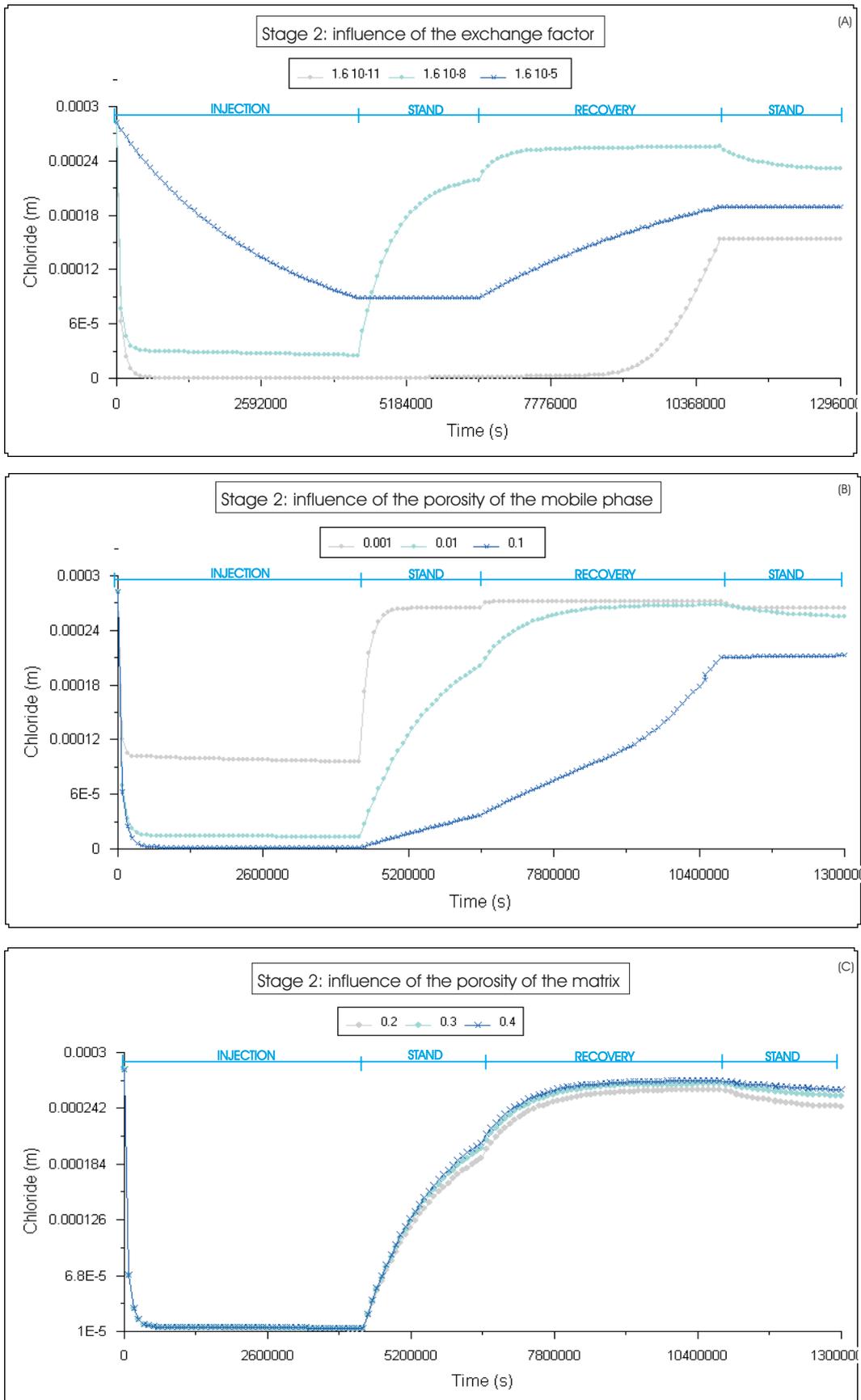
**Figure 7** Sensitivity of the permeability, layer thickness and porosity during ASR-operational cycles in the Lower Greensand



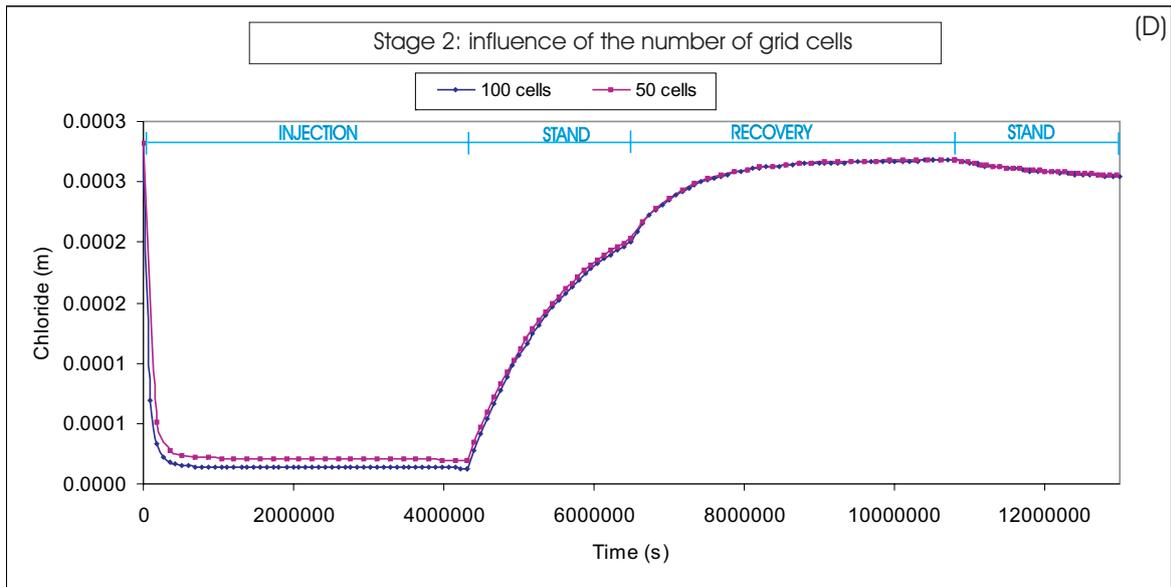
**Figure 8** ASR concentration curves for the four major aquifers



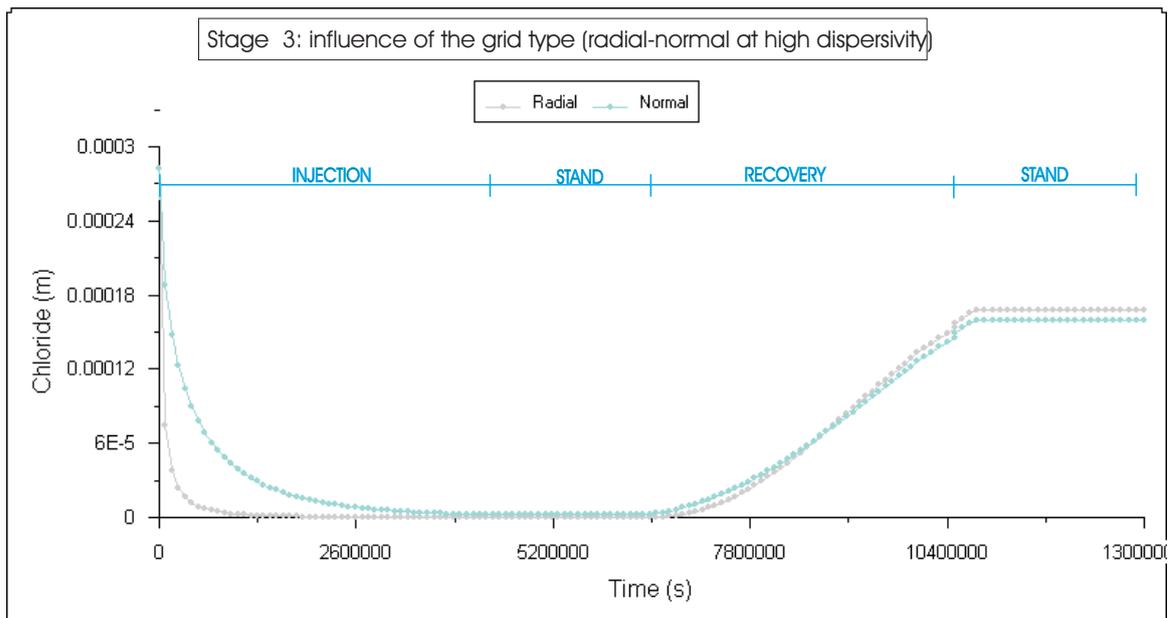
**Figure 9.** Stage 1: Influence of diffusion and dispersion on the modelled concentrations



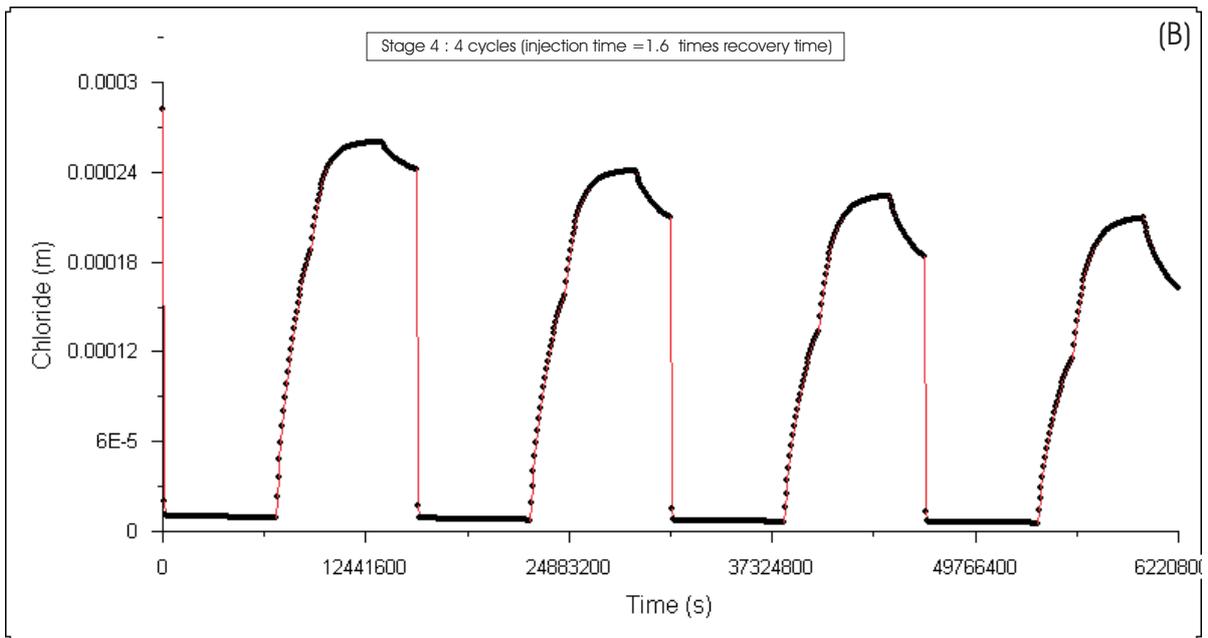
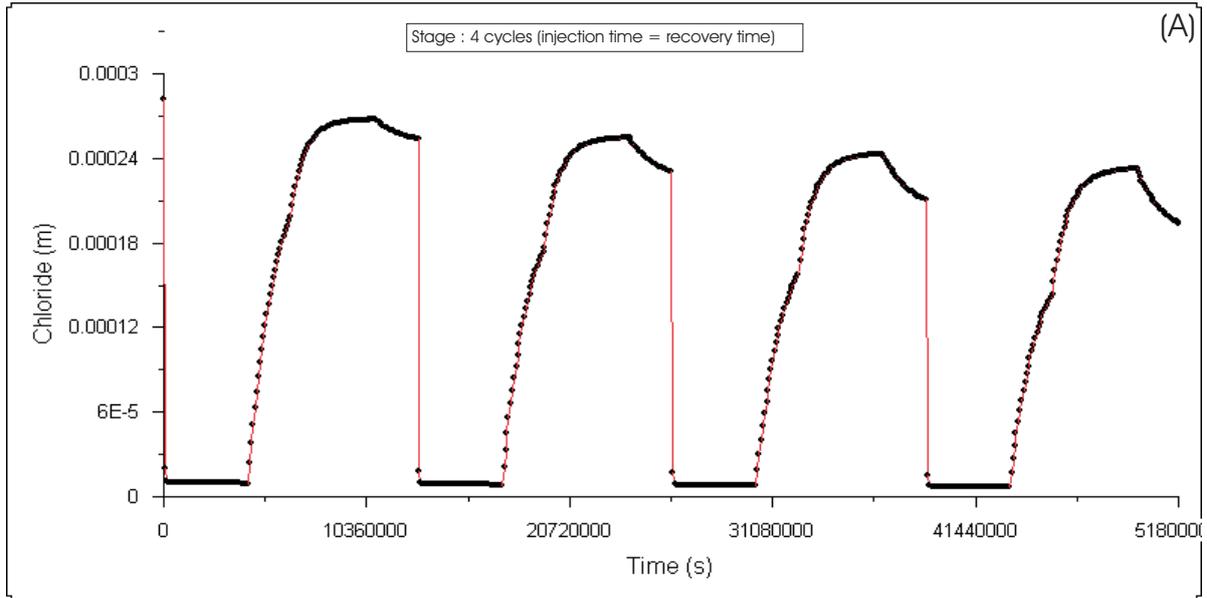
**Figure 10** Stage 2: Influence of the dual porosity parameters and the number of grid cells



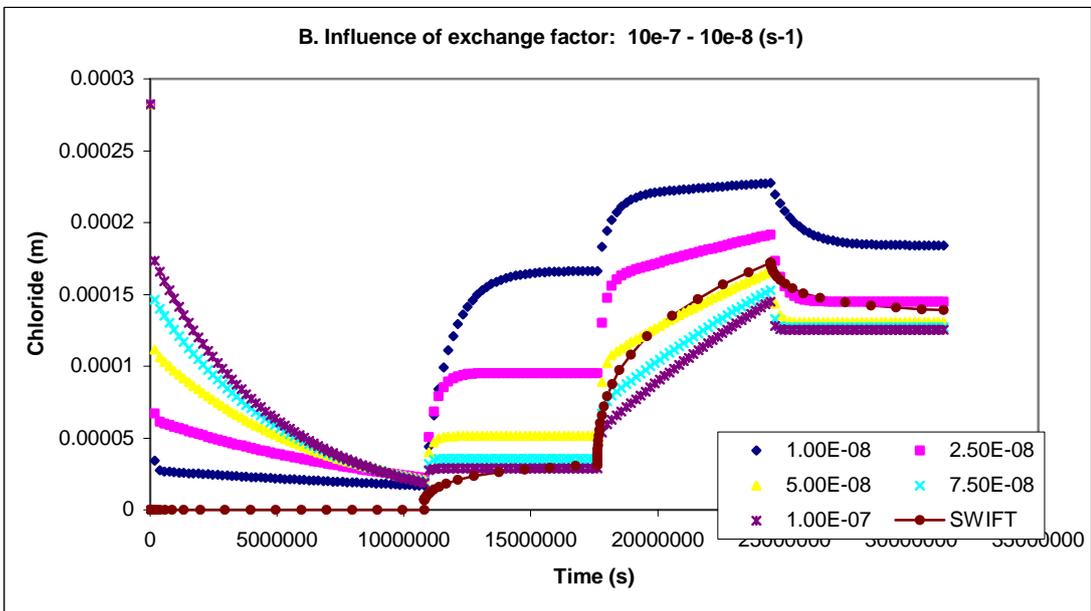
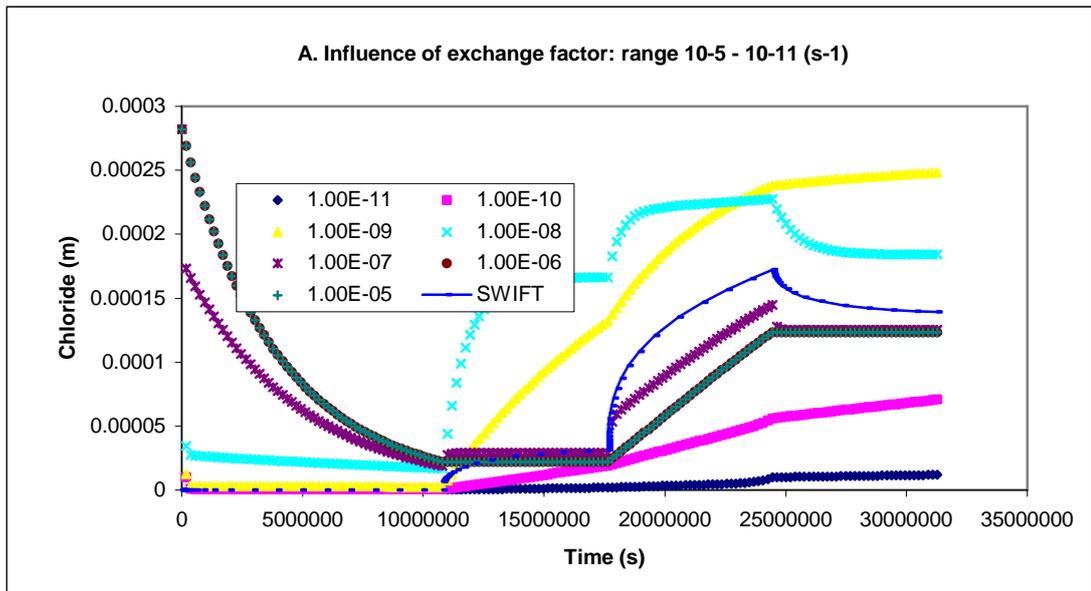
**Figure 10** Stage 2: Influence of the dual porosity parameters and the number of grid cells (continued)



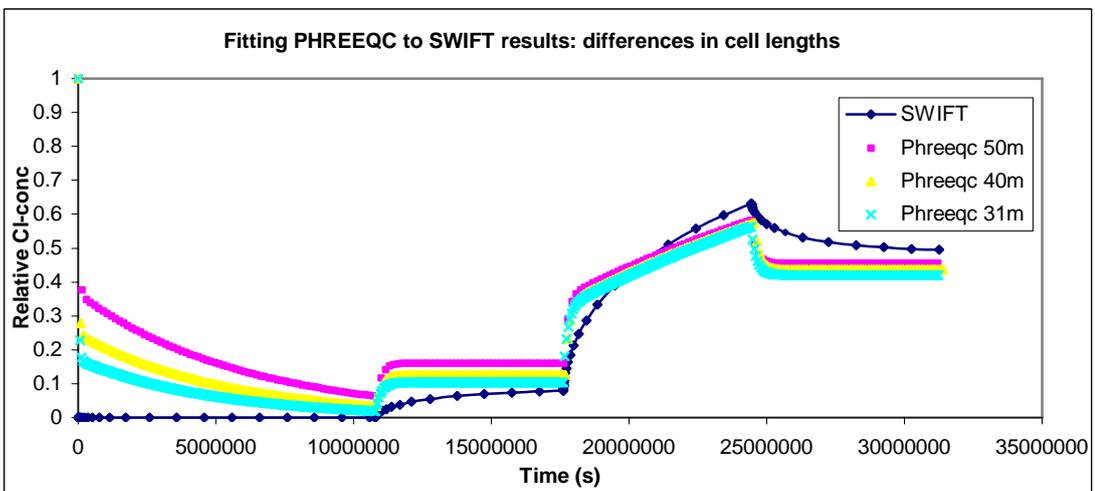
**Figure 11** Stage 3: Influence of the grid type (radial-normal)



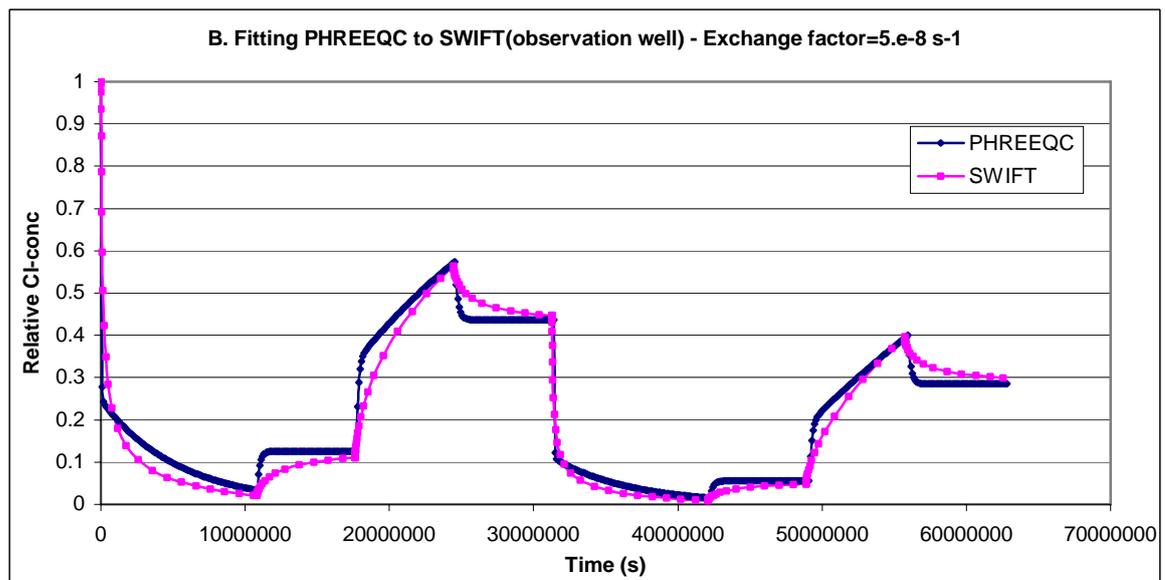
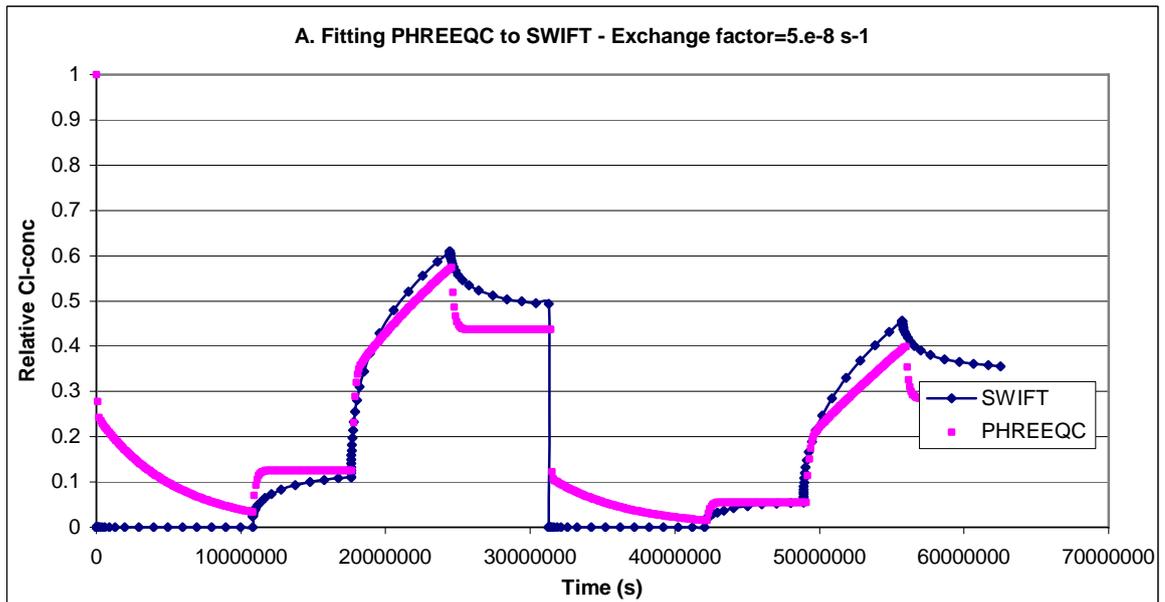
**Figure 12** Stage 4: Influence of different injection/abstraction ratios during multiple ASR-cycles



**Figure 13** Calibration of PHREEQC to SWIFT – fitting the exchange factor



**Figure 14** Calibration of PHREEQC to SWIFT – influence of the cell lengths



**Figure 15** Calibration of PHREEQC to SWIFT – optimised fit for the modelled concentrations in the well and at a distance of 20 from the well