Hydrological Characterization and Modeling of a Fractured Limestone Aquifer

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Abstract

An industrial site in eastern Denmark is contaminated with chlorinated solvents, mainly trichloroethylene (TCE). The TCE source is located in a fractured moraine till and it is leaking into an underlying fractured limestone aquifer. The contamination is currently being remediated using a pump-and-treat solution, which has successfully stopped the spread of TCE in the aquifer. However, the municipal goal is to replace the pump-and-treat solution with a more cost efficient remediation based on enhanced reductive dechlorination. This approach utilizes specific dechlorinating bacteria that are capable of degrading TCE under reduced conditions, created in the aquifer by injection of a substrate. To ensure optimal conditions for the enhanced reductive dechlorination, the new remediation system was designed using a local ground water model. The model was calibrated using hydrogeological characteristics of the limestone aquifer obtained through ground water well data. The hydrological characteristics were determined using a combination of water level measurements, borehole logging and well testing. Additionally, the transport characteristics of the limestone were determined by performing a tracer experiment with the fluorescent tracer Uranine. The results from the flow and transport characterization were used to calibrate the ground water model for the area in Visual Modflow. The model proved useful in designing the enhanced reductive dechlorination system. Specifically, the model revealed the optimal number and location of wells needed for injection of substrate as well as the necessary injection time needed for total reduction. The presentation will focus on the different methods used in characterizing the limestone aquifer and the inclusion of those results in the ground water model.

Introduction

Remediation of chlorinated solvents in fractured media constitutes one of the largest challenges within ground water remediation. Traditional pump-and-treat solutions are the most common remediation method in use, however because of the long time-scale involved it is very costly. New remediation techniques, like enhanced reductive dechlorination, are thus becoming increasingly popular for remediating chlorinated solvents in porous media [AFCEE, 2004]. This technique utilizes specific dechlorinating bacteria that are capable of degrading chlorinated solvents under reduced conditions, created in the aquifer by injection of a substrate. The technique has also been tested in fractured media [Chartrand et al., 2005], where the challenge is to ensure sufficient spreading of substrate and bacteria and to remediate the pollution in the matrix. To enhance the success of the method in fractured media, enhanced reductive dechlorination has recently been combined with environmental
fracturing to increase the spreading of substrate and bacteria in the otherwise low-permeable matrix [Scheutz et al., 2006].

In Denmark, fractured limestone is often used as a ground water reservoir, comprising a large part of the water resource. The limestone aquifers often underlay moraine till, where the fractures in the till render the limestone susceptible to pollution. Furthermore, characterizing flow and transport properties in the limestone is challenging given the heterogeneous character of the fractures. To obtain sufficient knowledge of the involved properties, several methods for accurate characterization are required. Using ground water models to examine processes in fractured media is likewise challenging. Consequently, several different modeling approaches exist from simple continuum models to more complicated discrete fracture network models. Selecting the most suitable model depends not only on the characteristics of the system being considered, but also on the availability of data and the purpose of the simulation.

The current study focuses on a trichloroethylene (TCE) pollution in a fractured limestone. The pollution is currently remediated with a pump-and-treat solution, which prevents spreading to a downstream water supply. The responsible municipal, however, wishes to replace the pump-and-treat solution with a more cost efficient remediation based on enhanced reductive dechlorination. To examine the possibilities of applying enhanced reductive dechlorination at the site, a ground water model for the limestone was calibrated and used to design the new remediation system. Model calibration was possible using data from an extensive test program designed to characterize the flow and transport properties of the limestone. The different methods used for that characterization are described herein, including application of the results in a local ground water model.

**Site description**

The study site is located in the town of Hellested in eastern Denmark. Geologically, the area consists of 9-10 m of fractured, clayey, moraine till with imbedded sand lenses overlaying a fractured limestone aquifer, described as a dense Bryozoan limestone with flint horizons imbedded.

Industrial production previously occurred at the site, resulting in pollution of chlorinated solvents, mainly TCE. The source of the pollution is located in the moraine till ~5 m below the surface. The pollution is slowly spreading to the underlying limestone aquifer and has been detected in a downstream water supply well approximately 200 m from the source. To prevent further spreading of TCE, a pump-and-treat solution was installed on the site in 1997. The pump-and-treat remediation takes place in three wells (218.1150, 218.1151, and 218.1152) at the site (Figure 1). The wells are 31-m deep with a 20-m casing in the limestone. Water is pumped from the downstream well (218.1150) at a rate of 4.5 m$^3$/h and after treatment it is re-circulated in the two upstream wells (218.1151 and 218.1152).

Additionally, seven new wells (K1-K7) were established as part of the current study. The purpose of the new wells was to examine the distribution of chlorinated solvents and use the
boreholes for characterizing the hydrogeology of the limestone aquifer. The wells were all established as open wells in the limestone, with a liner through the moraine till. Two wells, K2 and K7, extend 7 m into the limestone, whereas the remaining wells are 5 m into the limestone. Core drilling was used for well K2, which allowed a core to be extracted for visual inspection and laboratory experiments. The remaining wells were all drilled using the DTH (down the hole) method.

Figure 1. Location of existing and new wells.

Methods

Flow and transport properties of the limestone were characterized through different borehole data from the wells. Flow characteristics were determined through a combination of water level measurements, borehole logging, and a pumping test. Additionally, transport characteristics were determined through tracer test and level-specific water sampling.

Flow characterization

Water level was measured in the wells to determine the equipotential of the limestone aquifer. Measurements were made for two different situations: (1) without the pump-and-treat remediation running, thus providing the natural equipotential of the site and (2) with the pump-and-treat remediation running, thus providing a forced equipotential where water is being circulated through the aquifer at 4.5 m³/h.

Borehole logging was carried out in multiple wells on the site in order to determine the zones in which inflow to the wells was occurring. The logs included a televiewer log, gamma-ray log, induction log, temperature log, conductivity log, caliber log, flow log, and heat-pulse flow log. The limestone geology was visually viewed with the televiewer log and interpreted through the gamma-ray and induction logs. The remaining logs were used to
evaluate the hydraulic active zones in the limestone. The new wells (K1-K7) all had a relative low yield, which unfortunately made it impossible to perform the flow log. Instead, the flow pattern was interpreted using the heat-pulse flow log. Thus, the flow log was only performed in the deep pumping well 218.1150.

A short constant-rate pumping test was performed in well K7. The water level change was monitored using automatic loggers both in the pumping well and in the surrounding monitoring wells (K1, K3, K5, and K6). The pumping test was performed with a relative low yield (840 l/h) because the wells low conductivity. Pumping was continued for 3 hours followed by 24-hour measurements of the water level recovery.

Transport characterization

The transport characterization of the limestone aquifer was evaluated through a tracer experiment between the two wells K7 and 218.1150. The fluorescent dye Uranine was applied as a tracer since it is generally considered to be conservative in limestone material [Käss, 1998]. Furthermore, Uranine has an advantage compared to other conservative tracers as it can be easily monitored in the field with a fluorometer, thus reducing the required labor for sampling and analysis. The monitoring was accomplished using a flow-through fluorometer (GGUN-FL02, Geomagnetism Group, University of Neuchâtel, Switzerland), which continuously (every 10-minute) measured the Uranine concentration at a detection level of 0.02 ppb. The tracer experiment was performed synchronously with the pump-and-treat remediation, which ensured constant flow conditions in the limestone aquifer. The Uranine tracer was added as a short pulse (100 L) in well K7 at a concentration of 500 ppb. Afterwards, the concentration was monitored continuously in the pumping well 218.1150, located 34 meter downstream from the injection well, for a period of 60 days.

From each well, level-specific water sampling were performed for analysis of chlorinated solvents and redox parameters. Two different methods were used to extract the water samples: (1) using an inflatable packer system to isolate the well in 1 meter intervals for extracting local water samples and (2) to extract water samples during the borehole log using separation pumping. The results from the analysis will not be presented in the current presentation; however, the two methods will shortly be compared.

Results

Flow characterization

The hydraulic head measured in the wells, both during natural flow conditions and during pump-and-treat remediation, are shown as equipotential lines in Figure 2. In both cases, the flow direction is north-west. The natural gradient on the location is approximately 0.0125 m/m, however, during the pump-and-treat remediation, the gradient increase to 0.058 m/m.
The borehole logs reveal that the limestone is 9.5-11.5 m below surface. The televiewer log reveals that the limestone is dense, with embedded flint horizons up to 10 cm thick and no visual fractures. The heat flow logs performed in the shorter wells show that the majority of the inflow occurs in the top 1 meter of the limestone. Interpretation of the flow log performed in the deep well (218.1150) suggests that inflow takes place in five different horizons: 12% of the water flows in at the top of the limestone, 40% flows in at 0.6 m below limestone surface (m b.l.s.), 8% flows in at 3.9 m b.l.s., 8% flows in at 8.1 m b.l.s., and 22% flows in at 11.7 m b.l.s. Thus, the majority of inflow takes place within the top meter of the limestone. However, a significant part of inflow is also seen deeper in the well at 11-12 m b.l.s.

Drawdown and recovery of the water table due to the test pumping in well K7 is shown in a semi-log plot (Figure 3). After 10 minutes of pumping, a near-linear drawdown is seen in both the pumping and observation wells. The observation wells K1, K3 and K6 all show very similar drawdown, despite their difference in distance to the pumping well. In well K5, less drawdown is observed compared to the other wells. K5 is located diagonal to the other
observation wells, suggesting the existence of a dominant north-west flow direction, likely controlled by fractures in the limestone. Wells located on this dominant direction (K1, K3, and K6) should thereby show the same drawdown, whereas wells located diagonal to the flow direction will show less drawdown. Unfortunately, no well other than K5 is located diagonal to the suggested dominant flow direction to confirm this theory.

The measured drawdown and recovery data were analyzed with the program AQTESOLVE by Hydrosolve Inc. The best curve fit was found with a standard Theis solution, which indicates that the limestone can be described as an equivalent porous media (i.e. the fractures are not directly described in the model). Since the interpreted hydraulic values are going to be used as input in the ground water model for the site, which presumes that flow takes place in an equivalent porous media, the Theis solution is found to be satisfactory. The interpreted hydraulic values are between 1.4E-5 to 2.0E-5 m/s.

![Distance between wells:](image)

- K1-K7 12.5 m
- K3-K7 10.9 m
- K6-K7 8.4 m
- K5-K7 12.5 m

Figure 3. Measured drawdown and recovery from pumping test in well K7

*Transport characterization*

The breakthrough curve for the fluorescent tracer Uranine measured in the pumping well (218.1150) is shown in Figure 4A. The first tracer is detected after ~100 hours, followed by a sharp increase in concentration. Two peaks are detected at 200 and 375 hours, corresponding to 1.8% and 2.2% of the input concentration. The reason for the two peaks is unknown, though they may represent natural variation in the fracture system, variation in the mixing and thereby source concentration in the injection well K7, or different outer conditions such as variation in the pumping flow. The peaks are followed by a long tailing and at the end of the experiment (60 days after injection) the concentration was still elevated. Long tailing is typically seen in fractured media and is normally subscribed to matrix diffusion. However, in the current experiment, the dilution of tracer in well K7 might have added considerably to the tailing as well.
The accumulated tracer breakthrough curve 60 days after the injection is 58% of the tracer mass found in the pumping well (Figure 4B). If the experiment had continued longer, the significant tailing would have added to an even higher mass recovery.

![Breakthrough curve](image)

Figure 4. (A) Breakthrough curve for uranine measured in pumping well 218.1150 presented both as absolute and relative concentration. (B) Breakthrough curve showing relative accumulated mass.

Level-specific water sampling was performed at all wells for analyses of chlorinated solvents and redox parameters. In the deep pumping well, water was sampled during the borehole logging using separation pumping and analyzed for chlorinated solvents and Uranine, with the sampling occurring immediately after the tracer test ended. The water samples showed a surprisingly high concentration of chlorinated solvents in the lower part of the well, which suggests that the pump located at the bottom of the well, was pulling the pollution down into the aquifer. Analysis of the tracer Uranine confirmed this theory, since surprisingly high concentrations of Uranine also were detected at the bottom of the well.
Water sampling from the other wells were done either during borehole logging or using an inflatable packer. However, the results from the water samples taken using the two different methods did not show the same tendency; i.e. very different pollution levels were found in wells located close to each other. This suggests an inconsistency in the two methods, but it is unfortunately not possible to evaluate which of the methods are most reliable based on the available results.

Ground water model

A simple ground water model for the site was calibrated according to the characterization of the limestone. The model was constructed in Visual Modflow 4.0 with MT3D as the transport module.

The chosen model area was 500 x 1000 m (Figure 5). A simple two-layer geological model was constructed, with moraine till overlying 30 m of limestone. The limestone was divided into four horizontal zones based on borehole logging results. The top 4 m of the limestone was designated with the highest hydraulic conductivity, followed by 6 m with a low hydraulic conductivity, and 5 m with an intermediate hydraulic conductivity. The lower 15 m of the limestone was designated with a very low hydraulic conductivity.

The flow model was calibrated by changing the hydraulic conductivities for the various limestone layers. The data for the flow calibration included the water level measurement, both during natural conditions and during the pump-and-treat remediation, and the pumping test results, where the latter also was used for calibrating the transient flow model. The transport model was calibrated by changing the porosity and dispersion for the limestone layers to fit the results from the tracer experiment.

Figure 5. Model area for ground water model (500 m x 1000 m).
Flow model

By using the calibrated flow model, the water level during the natural flow condition and during the pump-and-treat remediation was simulated, Figure 6. The simulated heads match the water level measured in the wells satisfactorily.

Figure 6. Simulated equipotential lines during (A) natural conditions and (B) pump-and-treat remediation. The boxes show the comparison between measured and simulated heads.
Examples of simulated and measured drawdown and recovery during the pumping test in K7 are shown in Figure 7. Note that the same excellent fit is found for all wells. Based on these results, it is concluded that the model describes the flow in the limestone satisfactorily.

![Figure 7. Examples of measured and simulated water level during test pumping in well K7.](image)

**Transport model**

The applied transport module only simulates transport in porous media. Thus, for the model to be applicable, transport in the fractured limestone was assumed to occur in an equivalent porous media where fractures are not directly accounted for. This is a valid approach if the fractured media has a high and homogeneous fracture density (in contrast to a few dominant fractures) and a high permeability matrix [Wu, 2000]. For the limestone at the site, no significant fractures have been detected and the matrix is expected to be highly permeable.

Using the equivalent porous media approach, transport is simulated by the two parameters porosity and dispersion. Simulation of the tracer experiment with different values for porosity and dispersion is shown in Figure 8 and compared with the measured breakthrough curve. For a small porosity (n=0.13), which is expected in fractured limestone, the simulation captures the fast tracer breakthrough, though the tracer peak is over estimated. Increasing porosity lowers the simulated peak concentration, but results in a longer breakthrough time. This is overcome by increasing the dispersion to ensure earlier initial breakthrough and a longer tailing. The best simulated fit of the tracer experiment were thus found using a relative high porosity of 0.2 combined with a dispersion of 3 m.

An alternative solution to simulate the tracer experiment would involve using a more sophisticated model that describes the transport in fractures and matrix. The first step would be to use a double-porosity or double-permeability model to include the matrix diffusion component, followed by a discrete fracture model. However, with increasing complexity, the
number of parameters will also increase, thus requiring more calibration data (e.g. multiple tracer experiment). Given the purpose of the current model (i.e. the design of a new remediation system based on enhanced reductive dechlorination), a model based on an equivalent porous media is considered sufficient.

![Tracer experiment graph](image)

**Figure 8.** Modeling the tracer experiment using different values for porosity \( n [-] \) and dispersion \( D [m] \).

### Design of enhanced reductive dechlorination system

The calibrated ground water model was used to design a new remediation system for the site based on enhanced reductive dechlorination. The remediation technique takes advantage of specific dechlorinating bacteria which are capable of degrading TCE to non-chlorinated compounds (ethene/ethane) at reduced conditions. This is achieved by adding a substrate to the aquifer, which works as an electron donor, creating reduced conditions. The spreading of substrate and bacteria in the aquifer is thus essential for successful degradation of TCE.

Using the calibrated ground water model, different designs for injecting the substrate were simulated. Important parameters that were examined include: (1) number of injection wells, (2) injection and extraction rate, (3) substrate concentration, and (4) necessary injection time. The final design of the remediation system will not be described in detail in the current presentation, though examples of two different scenarios are shown in Figure 9.
Discussion and Conclusions

The characterization of the limestone took place in different types of wells. The new wells on the site were drilled using either core drilling or the DTH method. The advantage of the core drill is that a smooth borehole is produced and core samples can be extracted for visual expectation and laboratory experiments. In contrast, the DTH wells do not allow extraction of sediment and the borehole is less smooth. The advantage of DTH wells is that they generally produce more water than core wells. The new wells were open and without liners. Consequently, less uncertainty existed when performing borehole logging and level specific water sampling, since the filters would not disturb the natural flow system in the limestone. Open boreholes are, however, only applicable in materials that are sustainable and structurally coherent.

The flow in the limestone was characterized by a combination of water level measurement, borehole logging, and pumping test. All methods contributed to the understanding of the flow system. On the current site, it was possible to determine the flow conditions for two different flow schemes, with and without the pump-and-treat system, which yielded two different data sets for calibration. The use of borehole logging on the site was less successful, since the short wells did not produce enough water to run a flow log. The pumping test contributed considerable to the understanding of the flow system. The pumping test was relatively short, and a longer experiment would be useful to examine the dynamics of the fractured system. Despite the short pumping period, the results were used to evaluate a hydraulic conductivity for the limestone and provide data for calibration of the transient flow model.

The transport characteristics of the limestone were based on the tracer experiment and level-specific water sampling. The tracer experiment with Uranine was essential for calibrating the transport module in the ground water model. Additionally, the tracer experiment assisted in the interpretation of the detected TCE concentrations. The main disadvantage of a tracer experiment is the long time necessary for monitoring. Using a tracer like Uranine, which can be measured automatically in the field, reduces the time needed for sampling and analyses. The spreading of pollution in the limestone was evaluated with level
specific water sampling from borehole logging and packers. Analyses of samples from the
two different methods were inconsistent and showed differences that could not be explained
on the basis of the measured data.

The limestone characteristics were used to calibrate a ground water model for the site. The model assumes that flow and transport takes place in an equivalent porous media and the fractures are not taken directly into account. The calibrated model was capable of simulating the observed flow and transport properties satisfactory. Thus, the model was evaluated as capable of simulating the new remediation system on the site using enhanced reductive dechlorination.

References


Biographical sketches

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