

NONISOTHERMAL MULTIPHASE MODELING AS AN INTEGRATION TOOL IN THE PLANNING, CONTROL AND OPTIMIZATION OF IN SITU SOIL REMEDIATION IN SÖDERKULLA, SOUTHERN FINLAND

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ABSTRACT

The T2VOC numerical simulator has been used as an integration tool in an in situ remediation project in which TCE (trichloroethylene) contaminated soil is treated with steam and air injection and vapor extraction. Nonisothermal multiphase modeling has been applied in all phases of the remediation life-cycle: in the iterative conceptualization of the migration of the contaminants and the associated effects of the treatment operations, in the planning of optimal system design and controlling the processes during the remediation, as well as in the integration of the information obtained from the field investigations during the different phases of the project. The modeling has proved to be a valuable tool in decision making, and especially helpful in studying a wide range of questions during the remediation project.

INTRODUCTION: CASE SÖDERKULLA

The soil of the Söderkulla Estate in Sibbo Municipality, Southern Finland, is contaminated with TCE (trichloroethylene) originating from a metal workshop that was operating in the area more than thirty years ago. The soil consists of a 3-4 m thick clay layer surrounded by permeable sand. The contaminants are mainly located in the lower parts of the clay layer above the groundwater level.

The results of the soil survey are presented in Figure 1. Soil concentrations of over 10 mg/kg were found in a circular area with a diameter of about 15-20 m, the highest concentration being 231 mg/kg. Based on interpolation, the total amount of TCE in this area was estimated to be about 110 kg. At the time of the soil survey the highest detected TCE concentrations in the underlying aquifer were 700 µg/l.

TCE is a so-called dense, non-aqueous phase liquid (DNAPL) that is denser than water and forms a separate "NAPL phase" in the soil. If the NAPL concentration in the soil is under the residual saturation value, it does not migrate. It can, however, serve as an important source of secondary contamination by partitioning into the soil gas phase through volatilization, and into the soil water through dissolution. It

can thus provide a continuous source of groundwater contamination (Domenico and Schwarz, 1990).

The remediation actions were focused on the most contaminated area. Because the area is a historical milieu and there is a protected building (an old cowshed) located partly on top of the most contaminated area, remediation by excavation was not possible with reasonable costs. In situ treatment was therefore selected. A remedial system was designed (Figure 2) in which air and steam are injected under the clay in order to thermally enhance the clean up by soil vapor extraction (SVE).

Soil vapor extraction is a widely used technology for the removal of chlorinated solvents from the vadose zone (Relander et al., 1999). The clean-up process can be enhanced by thermal remediation techniques such as steam injection, which has proved to be particularly effective due to the high energy injection rate and the ability of steam to displace pore fluids even in saturated soils (Udell, 1996).

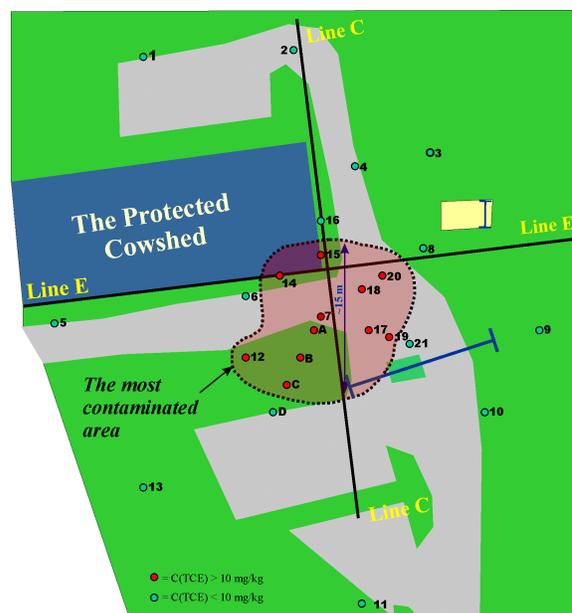


Figure 1. The results of the soil survey carried out in 2000 in Söderkulla.

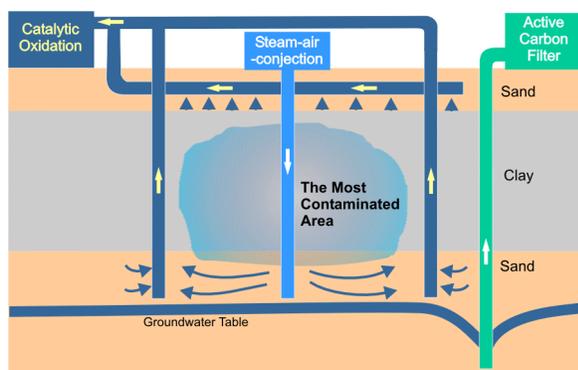


Figure 2. Process chart of the planned remediation system.

There are several mechanisms acting on the contaminants during steam injection: physical displacement, a decrease in the capillary and interfacial forces between the fluids and the porous media, enhanced desorption of contaminants from the porous solids, a decrease in the viscosity of the organic phase, steam distillation and steam displacement (Davis, 1998). When a liquid is present that is immiscible with water, the boiling point of the mixture is reached at a lower temperature than the normal boiling point of either of the liquids alone (Atkins, 1986).

Itamura and Udell (1995) have shown theoretically and numerically that depressurizing the steam zone by halting steam injection but continuing vapor extraction will cause a thermodynamically unstable system. To bring the temperature and pressure back into thermodynamic equilibrium, the temperature must be reduced to the point where it is in equilibrium with the reduced pressure of the system. Heat is lost by evaporation of the residual water and contaminants, which are then removed from the system by vacuum extraction. The injection of steam after equilibrium has been reached at a lower pressure allows recharge of the heat necessary to drive the evaporation of contaminants, and the depressurization cycle can then be repeated. (Davis 1998)

For sites in regions of sufficient permeability, steam injection and subsequent steam zone depressurization have been shown to be particularly effective in the removal of both the NAPL phase and aqueous phase contamination (Udell, 1998). Udell (1998) demonstrated using simulations that the clean up of lower permeability layers even can be considerable if they are surrounded by higher permeability areas. In his simulations, 90 % of the TCE was removed from an 8-m thick layer with permeability values of 10^{-12} , 10^{-14} and 10^{-16} m² in 1 hour, 4 days and 80 days, respectively.

PRELIMINARY SIMULATIONS

There were no data available about either the original TCE spill or the exact time of the spill. In the characterization of the contamination in the area and in the preliminary conceptualization process, the soil and groundwater data were combined with the information from a number of simulations.

If there is a spill of TCE near the ground surface, the contaminant percolates downward through the unsaturated zone towards the water table due to the potential gradient. In the unsaturated zone, the water present on the solid grains is the wetting phase and the nonaqueous phase liquid (NAPL) is the wetting phase with respect to air on the water film. Because water wets the solids, the NAPL does not displace the water from the surface, but instead moves from pore to pore once saturation exceeds the residual saturation. The NAPL displaces the pore water that is not strongly held, as well as the soil gas. If the spill is relatively small, downward percolation in the unsaturated zone will stop once the total volume is at residual saturation.

Due to capillary forces and varying hydraulic conductivity, the plume also exhibits a tendency to spread horizontally as it moves downward. Even a relatively thin, low permeability layer will inhibit downward percolation and force the contaminant to spread laterally. If the layer is discontinuous, the NAPL will eventually spill over and continue to move downward towards the water table. When NAPL finally reaches the capillary fringe, the pattern of spreading becomes more complex. Because water saturates a large proportion of the pores, the relative permeability to the immiscible liquid declines and, consequently, there is a tendency for NAPL to spread to some extent at the top of the capillary fringe (Domenico and Schwartz, 1990).

In the case of a DNAPL, the downward percolation continues in the saturated zone in a similar manner to that in the unsaturated zone. In this case, however, the water and not the soil gas is being displaced. The dense fluid moves to the base of the permeable unit as a result of driving forces related to the difference in density between the DNAPL and ground water. DNAPL accumulating on low permeability units will move downhill following the topography of the boundary. Spreading continues until the spill is at residual saturation (Domenico and Schwartz, 1990).

In Söderkulla the main geological feature influencing the spread of NAPL is the 3-4 m thick clay layer surrounded by permeable sand. According to the electrical tomography picture (Figure 3) that is combined with the soil sampling data, the clay layer (dark blue) is not continuous. Depending on the location of the spill, there are many possible preferential

flow paths on the top of the clay, through the clay or into the clay.

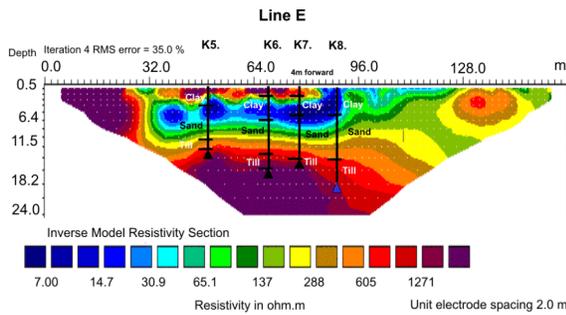


Figure 3. Electrical tomography picture in the form of a vertical cross-section of the Söderkulla site.

Spreading of TCE during an imaginary spill in this kind of heterogeneous soil with a clay block was simulated using a vertical 2-dimensional T2VOC-model (Falta et al., 1995, Pruess et al. 1999). In the simulations (Figure 4), NAPL (TCE) ponds on the top of the clay spread laterally, spill over, continue downwards, spread in the capillary fringe, continue through the saturated zone and pond at the bottom of the model. When the spill is stopped, the spreading continues until the total volume of TCE is at residual saturation.

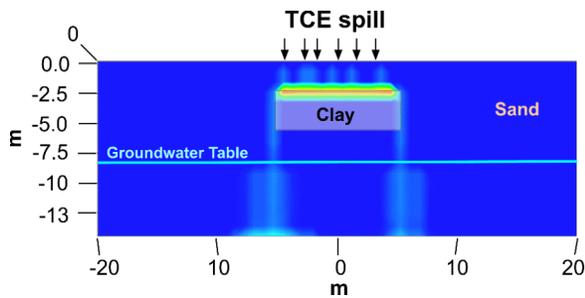


Figure 4. Simulated 2D-spreading of the NAPL phase in a vertical cross section during an imaginary spill of TCE.

In the simulation, no NAPL penetrated into the clay when the water content of the clay was about 35%. The annual precipitation was taken into account by assuming that 300mm/yr was infiltrated. In Söderkulla the detected water content of the clay in the soil samples was 33-44%. In the case of a long period of drying just before the spill, NAPL partly penetrated into the clay in the simulations. Matmon et al. (2001) have studied NAPL breakthrough during evaporative drying of clay barriers. They suggest that NAPL penetration and breakthrough occur as a dynamic process of micro-crack formation, rather than as a frontal NAPL advance. Crack formation is due to capillary effects on both NAPL-water inter-

faces and water-air interfaces. The upper edge of the clay in Söderkulla is near to the soil surface at a depth of 1 m. During a dry and warm summer period the clay may dry out in the upper parts, thus facilitating penetration of a possible NAPL spill into the clay.

The soil survey shows that the clay is not homogeneous, it has thinner parts, discontinuities and some digging has been done in the area; this means that preferential channeling-type spreading of NAPL into the clay is possible/probable. Ponding of rainwater and snowmelt has been reported on the top of the clay occasionally, but no permanent perched water has been found; this means that, if the contaminant spill has been large, occasional ponding of TCE on the top of the clay has taken place. TCE has dissolved in the soil water and volatilized into the soil gas from the NAPL ponds on the clay and from the preferential flow paths. In clay where the advective velocities are small, diffusion becomes significant or even a dominant mechanism for mass transport. However, diffusive spreading in the aqueous phase cannot be simulated with T2VOC. The new code TMVOC (Pruess and Battistelli, 2002) provides this possibility.

When the simulation was continued for thirty years after the spill, "natural clean-up" of the hot spot area by secondary contamination mechanisms could be seen. In the beginning of this clean-up process, the highest concentrations were in the high permeability areas but, at the end of the period, these had been cleaned up and the rest of the contamination was mainly left in the low permeability areas. According to the soil survey, this is the case in Söderkulla at the present time: The highest concentrations were found in the lower parts of the clay layer (at depths of 3,5 - 4 m) and in the basal till above the bedrock (at depths of 14,3 - 14,5 m). In the thick sand layer between these deposits, there were only small amounts of TCE remaining; the highest concentrations were found immediately below the most contaminated clay area. Vertical 2-dimensional simulations were also carried out to study the convective flow of TCE-contaminated water from the clay to the underlying groundwater. This was mainly done to show the necessity of cleaning up the hot spot area.

A 0-dimensional batch-model was used to study the distribution of TCE between NAPL, aqueous, and gaseous phases under different temperatures and soil conditions (table 1). The highest observed soil TCE concentration in Söderkulla was 231mg/kg in clay (above ground water level), and 11,1 mg/kg in sand (below the ground water level). In these soil concentrations TCE is in the aqueous and gas phases, although there is most probably an NAPL phase in the clay near the most contaminated sampling points.

The fact that these TCE concentrations are below the solubility limit does not necessarily imply the absence of an NAPL phase. Because of formation heterogeneities, it is possible that the distribution of TCE may be relatively non-uniform (Adenekan et al., 1990). According to table 1, an NAPL phase would be present in the clay if the soil concentrations were slightly higher. In sand the soil concentration should be much higher than the observed one for an NAPL phase to be present.

Table 1. Appearance of an NAPL phase for different concentrations of TCE in different soil conditions (in the simulations).

Soil type /C(mg/kg) ¹	NAPL% ⁴	
	T=6°C	T=20°C
Clay ² /238mg/kg	0 %	0 %
Clay ² /298mg/kg	17,2 %	11,4 %
Sand ² /11,3mg/kg	0 %	0 %
Sand ² /56,8mg/kg	0,1 %	0 %
Sand ³ /48mg/kg	0 %	0 %
Sand ³ /480mg/kg	45 %	45 %

- 1) Soil concentration
- 2) Unsaturated conditions
- 3) Saturated conditions
- 4) Mass of NAPL phase per total mass of TCE

The effectiveness of different remediation techniques was compared theoretically by means of 3D-simulations. The active part of the element model and the TCE distribution used are presented in Figure 5. In the simulations the clay was at a depth of 1,2 - 3,6 m, and the ground water level at a depth of 4,8 m. The background temperature was 10°C and the steam temperature 100°C. Steam was injected in two vertical wells in the center at a rate of 200 kg/h in each well, and air at a rate of 170 kg/h in each well. After the first month the steam injection rate was decreased to 50 kg/h. The injection wells were surrounded by four vertical extraction wells and five horizontal extraction wells above the clay. The extraction pressure was -500mbar.

This simulation should not be seen as a prediction of the duration of the clean up in Söderkulla, but rather as a qualitative study to compare different remediation strategies. The results are presented in Figure 6. The most effective method is of course air-steam-coinjection; with this method 99,8% of the TCE was removed in 8 months. With steam injection without air, only 68,5% was removed during the same period.

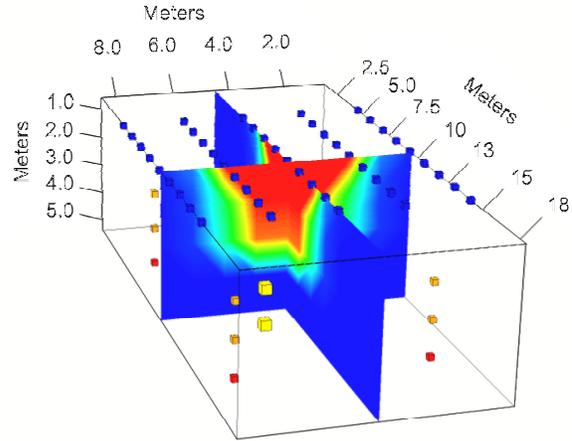


Figure 5. The active part of the model used in the preliminary remediation simulations. The soil TCE concentration in the red area is 250 mg/kg. The total amount of TCE is 50 kg

With a pure steam injection there is insufficient airflow to "carry out" the contaminants and, when the steam condenses, it causes contamination of the underlying groundwater. In this case the groundwater level is so close that, in a real remediation case, groundwater pumping should be employed. Air injection without steam seems to be relatively effective owing to drying of the clay caused by the dry injection air. When trying to conserve energy and, at the same time, to avoid drying the clay, air and steam can be injected cyclically. Itamura and Udell (1995) have shown that cycling steam injection with continuous vacuum extraction will always reduce the amount of steam required to meet a given clean-up level, and may also reduce the overall cleaning-up time.

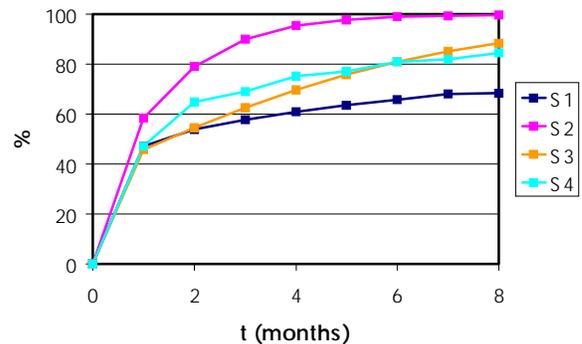


Figure 6. The proportion of extracted TCE (%) with different strategies in the simulations. S1: SVE + steam injection; S2: SVE + steam-air-coinjection; S3: SVE + air injection; S4: SVE + cyclical air and steam injection.

The soil parameters used in the simulations are presented in tables 2-4. The values were chosen on the basis of the literature (Mercer & Cohen, 1990; Pruess, 1992; Sundberg, 1988; Jääskeläinen et al., 1982), taking into account the Söderkulla site conditions.

Table 2. Soil parameters used.

	Sand	Clay
k	1e-11 m ²	1e-15 m ²
n	35 %	50-55 %
D _{rock}	2650 kg/m ³	2650 kg/m ³
SPHT	800 J/kg°C	800 J/kg°C
C _{wet}	1,5	0,8
C _{dry}	0,5	1,8

Table 3. Relative permeability function (Stone, 1970).

	Sand	Clay
S _{wr}	0,1	0,6
S _{nr}	0,05	0,05
S _{gr}	0,01	0,01
n	3	3

Table 4. Capillary pressure function (Parker et al., 1987).

	Sand	Clay
S _m	0,0	0,0
n	1,84	1,84
α _{gn}	5,0e4	5,0e4
α _{nw}	5,24	0,5

SIMULATIONS OF THE PILOT PHASE

A remedial system was installed in which air and steam are injected under the clay layer in order to heat the soil and enhance the clean-up effectiveness by soil vapor extraction (Figure 2). Contaminant vapors are treated by catalytic oxidation. Groundwater is pumped and treated in an active carbon filter. The remedial process is automated and can be controlled remotely.

During the pilot phase (22.2. - 31.5.2001), warm air was injected under the clay layer from two vertical wells in the center, and soil vapors were extracted from four vertical wells located around the injection wells and from five horizontal wells above the clay.

The system was planned to be closed according to in situ permeability tests.

Steam was first injected from the air injection wells (300 kg/h, 200 °C). After some time, however, the steam pressure cut through the clay and the injection had to be stopped. The problem was solved by replacing the steam generator with a smaller one (120 kg/h, 120 °C), and by installing new steam injection wells. During steam injection intensive monitoring was performed in order to collect data for model calibration and for designing a full-scale treatment system. Temperature was measured at 0,5 m intervals along nine vertical lines down to 8-m depth. The maximum temperature in the soil was 102,8 °C at a depth of 4,5-5,5 m. The maximum temperature at a distance of 5 meters from the injection wells was 82,3 °C.

The pilot phase remedial actions were simulated using the 3D-T2VOC-model shown in Figure 7. The model was based on the soil survey, the preliminary simulations, and the planned and installed remediation system. Throughout the pilot project all the new data obtained in the field were used to make the model more closely resemble the real situation. The layer thicknesses and the groundwater level, as well as the initial and boundary conditions, were modified until the model functioned correctly and there was good agreement with the structural and functional data.

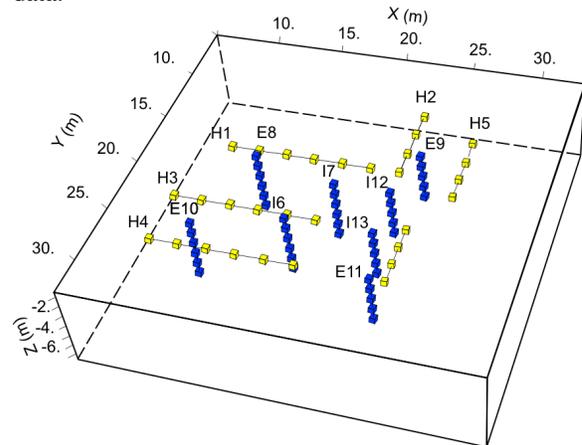


Figure 7. The active part of the simulation model used for the pilot scale simulations. Injection wells I6, I7, I12 and I13. Extraction wells E8-E11 (vertical), H1-H5 (horizontal).

The groundwater layer was described by three element layers and forced to the level measured in the standpipes. The upper boundary was assumed to be closed because the site was covered with an airtight covering (this assumption was later changed to allow heat flow through the boundary).

The initial soil conditions were calculated using simulations with an annual infiltration of 300 mm (half of the annual precipitation). With these assumptions and the parameter values presented in Tables 2-4, the simulated soil moisture content was in good agreement with the soil moisture content distribution (Figure 8) measured using a radiometric in situ method.

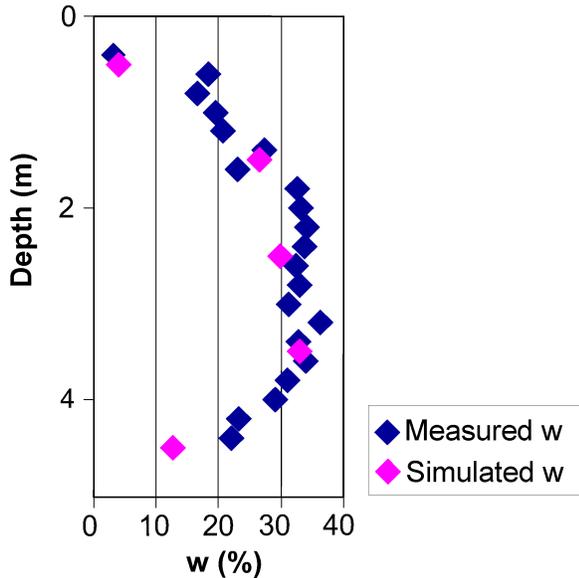


Figure 8. Simulated and measured soil moisture content w . (Clay at a depth of 1 - 4 m, groundwater level at a depth of 5 m.)

The other boundaries were handled by inactive elements at a distance outside the radius of the influence of the wells, and the background temperature was assumed to be 7°C , which is the average yearly temperature in Southern Finland. The final calibration was made according to the temperature measurements to give an agreement with 5°C tolerance. The breakthrough of the clay by the steam was described by changing the permeability of the clay around the injection wells to a higher value, $k = 10^{-11}$.

In the simulations several approaches were used to describe the contamination. To obtain information about the clean-up effectiveness in the area, a uniformly distributed degree of contamination was used in the whole area; in order to obtain a more realistic picture about the situation, a concentration distribution was used in which the local concentrations were interpolated from the soil sample data.

In the simulations, that are presented in Figures 9 and 10, the target area was divided into two parts: a slightly contaminated area (18m x 18m x 5m) with a soil concentration of 25 mg/kg, and a highly contaminated area (10m x 10 m 3 m) in the middle

with a soil concentration of 225 mg/kg. The total initial amount of TCE was 156 kg. In the simulations the influence of implemented clean-up operations focused on the middle and the northern side of the area, and the southern parts of the area were left almost untouched.

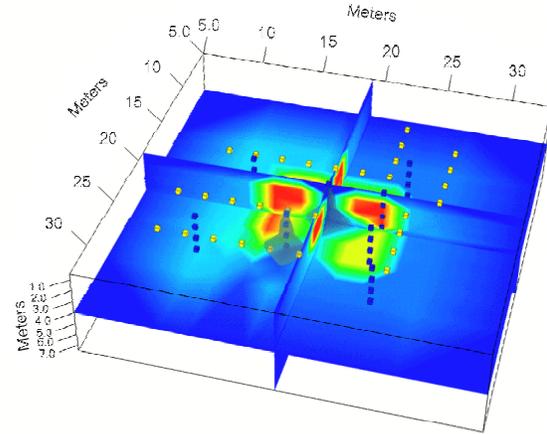


Figure 9. Simulation: Steam breaks through the clay around injection wells 16 and 17. In the gray area, $T > 60^{\circ}\text{C}$.

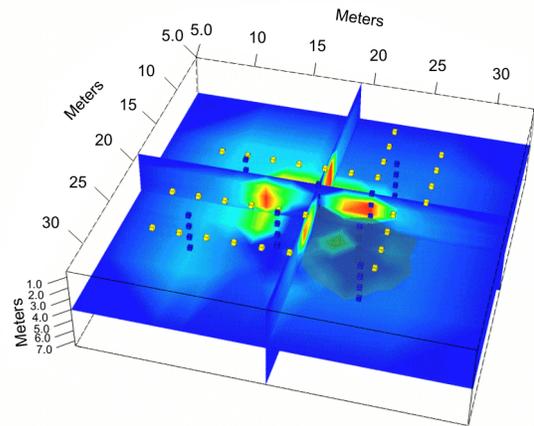


Figure 10. Simulation: The new injection wells 112 and 113 are in use. In the gray area, $T > 60^{\circ}\text{C}$.

The simulated amount of TCE extracted during the clean up was compared to measurements based on the temperature difference in the catalytic oxidation process. During the first two months the agreement was good: in the simulations 44 kg of TCE was removed, while according to the measurements the minimum amount of TCE extracted was 43 kg. As the effect of cooling on the catalytic oxidation process was not taken into account in this minimum value, the real value should be larger. On the other hand, it was not possible to take all the components that were oxidized in the process into account in the monitoring calculations. This would suggest that the

real value is smaller than the measured one. In the full-scale project the monitoring based on the catalytic oxidation process is calibrated with vapor samples.

At the end of the pilot project the total amount of TCE extracted was estimated to be 100-150 kg, which was more than the original estimate of the total amount of contaminant in the soil. This degree of extraction could be achieved with simulations in which the original amount was 300 kg. This example illustrates the difficulty in predicting the remediation efficiency. In the case of an NAPL chemical, the contaminant distribution in the soil tends to be highly heterogeneous, which makes it extremely difficult to determine the total amount of contaminant initially in the soil. If we do not know the initial contaminant distribution it is impossible to predict the clean up time. The power of the modeling lies in its ability to compare the effect of different strategies or measures on the clean-up efficiency, or in studying the associated effects or risks during the treatments

Drying of the clay is one risk that should receive attention in the case where a house is standing on the clay. The drying effect was studied with simulations by varying the injection-extraction combinations and following the effects. In the simulations, the effect of soil vapor extraction without air injection was minor, and only low drying effects were seen in the immediate surroundings of the wells. With air injection, on the other hand, drying occurred throughout the whole soil matrix between the wells, depending of course on the dryness of the air. With totally dry air, the soil moisture content of the clay was lowered after 8 months by 13% in the 1-m thick bottom layer of the elements and by 7 % in the next layer. With air-steam co-injection some drying occurred when the temperatures were at a maximum but, after steaming, the soil moisture content soon reverted to the original value due to the effects of steam condensation. Because of the possible risks of the clay drying during long periods of pure air injection, a radiometric in situ measurement of the soil moisture content was taken into use near the foundation of the cowshed.

The model was used in planning a full-scale treatment system capable of heating the desired area. The planned full scale system, and the simulated temperature distribution along the lower edge of the clay after two weeks of steaming, are presented in Figure 11. Two new injection wells (I14 and I15) were needed to heat the whole area, and two new vertical extraction wells (E16 and E17) for effectively collecting the contaminant vapors. The groundwater level had to be lowered by 0,5-1 m.

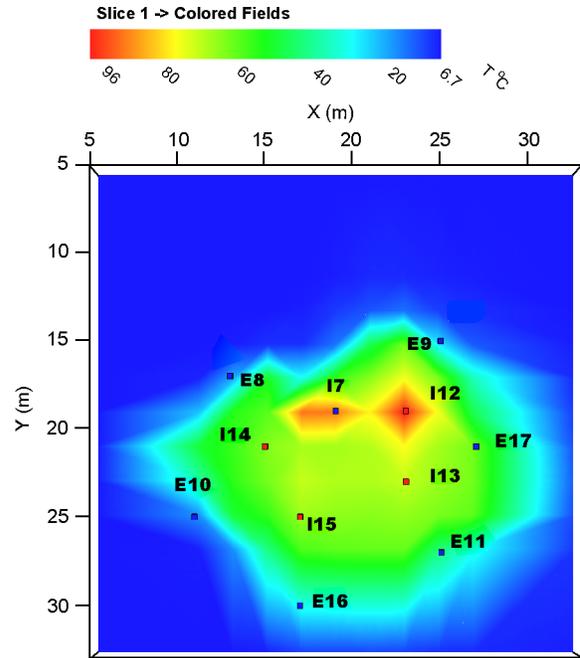


Figure 11. The planned full-scale remediation system and a simulated temperature distribution along the lower edge of the clay after two weeks of cyclical-rotational steaming from wells I12-15. In these simulations the former injection well I7 was used for vapor extraction.

FULL-SCALE REMEDIATION

The full-scale remediation system has been operating since 20.11.2001. After the pilot phase, new injection and extraction wells (I14, I15, E16, and E17) were installed in the area. Two new groundwater pumping wells have also been installed on the southern side of the area. The ground water level has been lowered by about 1 m, and some wells (I6, I7, E8 and E10) have been deepened. Temperature monitoring is carried out along 10 vertical lines, and pressure monitoring at 6 points under the clay and 2 points above it. Radiometric in situ measurement of the soil moisture content is carried out in one well near the cowshed, and the groundwater level and contamination is monitored in 8 standpipes.

For steam injection there are two steam generators: a small one (20 kg/h) that is used for steam-air co-injection, and a larger one (360 kg/h) for cyclical steam injection that is distributed to 2-4 injection wells at a time. The extracted gases are handled by catalytic oxidation, and the pumped groundwater with an active carbon filter.

The model has been updated and different injection-extraction combinations, cycles and rotations are compared with the simulations in order to find the most cost-effective and time-saving strategies. The model used for the simulations is presented in Figure 12, and some simulation results in Figure 13.

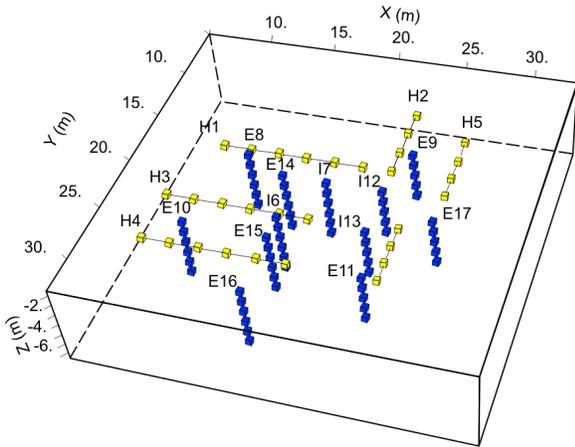


Figure 12. The active part of the model used for the full-scale simulations. Injection wells I6, I7, I12, I13, I14 and I15. Extraction wells E8, E9, E10, E11, E16 and E17.

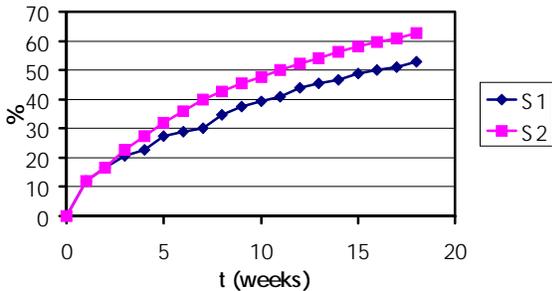


Figure 13. Proportion of TCE (%) extracted with different strategies in the simulations. S1: Cyclical steam and air injection. S2: Rotational steam injection with constant air injection.

In the simulations the most time-saving strategies seem to be those in which steam injection is made rotationally from wells I12, I13, I14 and I15, while keeping air injection on all the time in the middle (I7) by means of a small steam co-injection. The more frequently the steam injection points are changed, the higher is the extraction rate.

In the real situation it has not been possible to follow the planned steaming strategies rigorously. There

have been delays in getting the system to work properly, and with high levels of steam injection somebody has to be present at the site to take care of the steam generator and to control the system. This means that the active operations (changing injection wells, using a high steam injection rate) are limited to working hours. In the future it is hoped that these operations can be partly automated, and the operational scheme (pumping volumes and schedule, steam temperature and injection locations) further optimized by means of optimization techniques (Finsterle and Pruess, 1994) to reduce the remediation costs.

CONCLUSIONS

Simulations can be used as an integration tool in the planning, control and optimization of in situ treatments. Modeling can be applied in all the phases of the remediation life cycle. Simulation can be helpful in providing a more realistic picture of the contamination, a more illustrative view of the remediation process, and more concrete alternatives for decision-making situations. It can also be helpful in minimizing the risks, and maximizing the efficiency of the clean-up process.

It would be useful to adopt modeling as one of the approaches already in the site identification phase. The modeling point of view can broaden the understanding of the case, and lead to savings in other methods like soil sampling. There is always a delay between starting the modeling and receiving the results, but having the modeling as one of the tools and developing the modeling approaches iteratively, together with traditional site investigations, would be useful for both sides.

The modeling approach can be used to simulate the migration of contaminants and the clean up with a specific method under the conditions prevailing at the site, and the effect of different solutions on the costs and the duration of the clean-up project can be calculated. In some situations the simulations could give us hints about what is realistic and what is not, or expose systematic errors in the measurements. Modeling is an integrative tool that makes it possible to handle the interconnected effects of the processes.

It is, however, extremely difficult to forecast the costs or the duration of the whole project. It is often said that "the models give a too optimistic picture about the efficiency of the method". It may be true that, in the planning phase, there is a tendency to make too optimistic predictions, but it is also true that the simulations do not include interruptions caused by factors other than the physical or chemical processes or the soil conditions; almost anything can happen in any project connected to the real world.

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