Direct and Inverse Modeling of Multiphase Flow Systems

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SUMMARY

A modeling study is presented which demonstrates how the combination of simulation and optimization techniques can be used to improve the design of a multi-component remediation system. A series of computer codes has been developed at the Lawrence Berkeley National Laboratory to solve forward and inverse problems in groundwater hydrology. Simulations of non-isothermal, three-phase flow of volatile organic compounds in three-dimensional heterogeneous media were performed. Inverse modeling capabilities have been developed which can be used for both automatic model calibration and optimization of remediation schemes. In this study, we discuss a sequence of simulations to demonstrate the potential use of numerical models to design and analyze cleanup of a contaminated aquifer.

INTRODUCTION

Industrial processes involving metallurgical operations frequently cause contamination of soil and groundwater due to the release of solvents such as chlorinated hydrocarbons and other volatile organic compounds (VOCs). In the subsurface, these chemicals tend to remain as nonaqueous phase liquids (NAPL), and are transported under multiphase flow conditions due to gravitational, viscous and capillary forces. Other physiochemical processes also taking place include evaporation into the soil gas phase and dissolution into the groundwater. In the gaseous phase, VOCs are transported by multi-component diffusion in addition to density and pressure driven advection. In the liquid phase, dissolved contaminants may cause density effects which affect advective transport. Simulation of contaminant behavior in the subsurface requires accounting for all these mechanisms. Accurate process description and robust implementation of multiphase flow in porous media including phase changes are essential for the modeling of air sparging, steam flooding, and vapor extraction as some of the most effective remediation technologies such as air sparging, steam flooding, etc.

The design of a cleanup operation for a contaminated aquifer poses problems of a hydrological, technical, environmental, and economic nature. Decisions have to be made regarding aquifer testing for site characterization, the design of a containment system, the choice of an appropriate remediation technology, which usually requires specifying an injection and pumping schedule, and the layout of a monitoring system. First, it is crucial to characterize the relevant hydrogeological features at a given site. Information about the system state under natural or test conditions have to be collected to calibrate the numerical model. Secondly, a remediation technology is chosen, and cleanup operations are simulated to improve the understanding of the fate of the contaminants in the subsurface. A comparison between alternative approaches is performed to design and optimize the remediation strategy.

Most of the tasks described above require solving some kind of optimization problem, i.e. the maximization or minimization of a performance measure. An incomplete list is given in Table 1. The testing and data collection program can be optimized to obtain as much information about the plume location and hydrogeologic properties of the aquifer as possible. The numerical model to be used for the subsequent studies has to be calibrated against the available data. Model calibration involves minimizing the difference between the observed and the calculated system state. It is important to realize that the result of the subsequent modeling studies as well as the conclusions drawn regarding the optimum remediation system design depend on the accuracy with which the relevant features at the site are represented in the numerical model. Once an appropriate remediation technology has been selected, operational details such as the layout of the containment system, temperature and rate of air or steam injection, the pumping schedule, etc. can be optimized to increase the effectiveness and efficiency of cleanup. This step requires the definition of a cost function which comprises actual operational costs as well as hypothetical costs for residual contamination. It is obvious that selection of the remediation technology and optimization of the operational parameters is an iterative process. The overall performance of the optimized remediation alternatives are compared to determine the most appropriate strategy. Finally, the design of a monitoring system is improved by evaluating the amount of information that can be obtained from a limited number of sensors.

Table 1 Optimization Problems in Aquifer Remediation

Objective	Task	Simulation Problem	Optimization Problem
data collection for plume mapping and site characterization	test design, data collection	simulate plume spreading and hydraulic tests	maximize sensitivity of data with respect to parameters of interest
site characterization	model calibration	simulate system state under natural or test conditions	minimize difference between measured and calculated system state
effectiveness, efficiency	technology selection, determination of operational parameters	simulate cleanup operation	minimize cost function comprising actual and hypothetical costs
control	design of monitoring system	simulate migration of residual contamination	maximize obtainable information

The modeling study presented in this paper aims at demonstrating how the combination of simulation and optimization techniques can be used to improve the design of a multi-component remediation system. Computer programs have been developed at the Lawrence Berkeley National Laboratory to solve forward and inverse problems in groundwater hydrology. Simulations of non-isothermal, three-phase flow of volatile organic compounds in three-dimensional heterogeneous porous media were performed. Inverse modeling capabilities have been developed which can be used for both automatic model calibration and optimization of remediation schemes. We first discuss the modeling approach used to simulate multiphase contaminant transport, before we provide some information about the optimization algorithms. A sequence of simulations of a hypothetical contaminated site demonstrates the potential use of numerical models to design and evaluate cleanup operations.

MODELING MULTIPHASE CONTAMINANT TRANSPORT

Sophisticated numerical simulators have been developed at the Lawrence Berkeley National Laboratory to model multiphase, multi-component flow and transport in the subsurface. The main computer code used in this study is T2VOC [1] for three-phase (gas, water, NAPL), non-isothermal flow of water, air, and a volatile organic compound (VOC) in three-dimensional heterogeneous porous media. T2VOC is an extension of the TOUGH2 general-purpose simulation program [2] which uses a general integral finite difference formulation for mass and energy balance equations. The balance equations for three mass components κ (κ = w-water, a-air, c-chemical) and heat (κ = h-heat) are written in integral form for some flow region, V_n, having a surface area Γ_n, as follows:

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_{V_n} M^{\kappa}\,\mathrm{d}V_n = \int_{\Gamma_n} \mathbf{F}^{\kappa} \bullet \mathbf{n}\,\mathrm{d}\Gamma_n + \int_{V_n} q^{\kappa}\,\mathrm{d}V_n \quad . \tag{1}$$

Here M^{κ} is the amount of component κ per unit porous medium volume, $\mathbf{F}^{\kappa}$ is the total flux of component κ into V_n, $\mathbf{n}$ is the inward unit normal vector on Γ_n, and q^{κ} is the generation rate per unit volume. The three fluid components air, water, and VOC may be present in different proportions in any of the three phases gas, aqueous, and NAPL. Thus, the mass accumulation terms (κ = w, a, c) contain a sum over the three phases β (β = g-gas, w-aqueous, n-NAPL):

$$M^{\kappa} = \phi \sum_{\beta} S_{\beta}\, \rho_{\beta}\, X_{\beta}^{\kappa} + \delta^{\kappa c}\, \rho_b\, \rho_w\, X_w^c\, K_D \quad . \tag{2}$$

Here ϕ is the porosity, S_{β} is the saturation occupied by phase β, ρ_{β} is β-phase density, and X_{β}^{κ} is the mass fraction of component κ in phase β. The organic chemical accumulation term (κ = c, $\delta^{\kappa c} = 1$) includes the effect of linear equilibrium adsorption onto the solid phase, where ρ_b is the dry bulk density of the soil, and K_D is the solid-aqueous distribution coefficient for the organic chemical [3]. The heat accumulation term (κ= h) includes contributions from both the solid and the fluid phases:

$$M^h = (1 - \phi)\, \rho_R\, C_R\, T + \phi \sum_{\beta} S_\beta\, \rho_\beta\, u_\beta \ , \tag{3}$$

where ρ_R is the soil grain density, C_R is the heat capacity of the soil grains, T is the temperature, and u_β is the specific internal energy of phase β.

Each phase flows in response to pressure and gravitational forces according to the multiphase version of Darcy's law which includes the effects of relative permeability and capillary pressure between the phases. The mass flux terms of component κ (κ= w, a, c) include a sum of contributions from the three phases β (β = g, w, n):

$$\mathbf{F}^\kappa = \sum_{\beta} \mathbf{F}_\beta^\kappa \ . \tag{4}$$

For aqueous and NAPL phases (β = w, n) we have $\mathbf{F}_\beta^\kappa = \mathrm{X}_\beta^\kappa\, \mathbf{F}_\beta$, where the phase fluxes $\mathbf{F}_\beta$ are given by

$$\mathbf{F}_\beta = -\mathrm{k} \frac{k_{r\beta}\, \rho_\beta}{\mu_\beta} \left(\nabla P_\beta - \rho_\beta\, \mathbf{g} \right) \ . \tag{5}$$

Here k is the absolute permeability, $k_{r\beta}$ is the relative permeability of phase β, μ_β is the β-phase dynamic viscosity, P_β is the fluid pressure in phase β, and $\mathbf{g}$ is the gravitational acceleration vector. Mass flux in the gas phase includes advection and multicomponent diffusion:

$$\mathbf{F}_g^\kappa = -\mathrm{k} \left(1 + \frac{b}{P_g} \right) \frac{k_{rg}\, \rho_g}{\mu_g} X_g^\kappa \left(\nabla P_g - \rho_g\, \mathbf{g} \right) + \mathbf{J}_g^\kappa \ . \tag{6}$$

Here b is the Klinkenberg factor [4], which accounts for increased effective gas permeability at low pressures, and $\mathbf{J}_g^\kappa$ is the diffusive mass flux of component κ in the gas phase. The diffusive flux is driven by a mass fraction gradient, ∇X_g^κ, and is a function of saturation, tortuosity, and the multicomponent molecular diffusion coefficient (for details see [1]). Aqueous and gas phase pressures are related by

$$P_w = P_g + P_{cgw} \ , \tag{7}$$

where $P_{cgw} < 0$ is the gas-water capillary pressure. The NAPL phase pressure is related to the gas phase pressure by

$$P_n = P_g + P_{cgn} \ , \tag{8}$$

where $P_{cgn} < 0$ is the gas-NAPL capillary pressure. Assuming the NAPL is the intermediately wettable phase, the NAPL-water capillary pressure, P_{cnw}, is given by

$$P_{cnw} = P_{cgw} + P_{cgn} = P_w + P_n \ . \tag{9}$$

The heat flux includes both conduction and convection

$$\mathbf{F}^h = - K \nabla T + \sum_{\beta} h_\beta \mathbf{F}_\beta \ , \tag{10}$$

where K is the overall porous medium thermal conductivity, h_β is the β-phase specific enthalpy, and $\mathbf{F}_\beta$ is the total mass flux of phase β.

Finally, the sink and source term in Eq. (1) includes external contributions, e.g. the release of contaminant or the injection of steam as well as the production of a three-phase fluid mixture during forced vacuum extraction. In addition, a sink term may be included that permits an approximate representation of biodegradation of VOCs (for details see [1]).

Water properties in the liquid and vapor state are calculated using steam table equations given by the International Formulation Committee [5]. Thermophysical properties of the NAPL phase such as saturated vapor pressure and viscosity are calculated as functions of temperature, while specific enthalpy and density are computed as functions of both temperature and pressure. Gas phase thermophysical properties including molecular diffusivities are considered to be functions of temperature, pressure, and gas phase composition. A general equation of state is provided to compute the necessary NAPL/VOC thermophysical and transport properties. Based on semi-empirical corresponding states methods, the chemical parameters are calculated as functions of critical properties such as the critical temperature and critical pressure, which are readily available for many substances (see for example [6]). This approach makes T2VOC applicable to a variety of contamination problems involving different NAPLs that are either denser or lighter as water, and that have different vapor pressures and solubilities.

For numerical solution, the balance equations (1) are discretized in space based on an integral finite difference formulation [7]. Time is discretized fully implicitly using first-order backward finite differences. Discretization results in a set of nonlinear coupled algebraic equations which are solved simultaneously by means of Newton-Raphson iterations. A conjugate gradient algorithm is used to solve the linear equations arising at each iteration step [8].

OPTIMIZATION ALGORITHM

As discussed in the introduction, we use the simulation capabilities in combination with optimization techniques to perform model calibration and to support the design of a remediation system. The first task is referred to as inverse modeling, whereas optimization of a remediation design is a prevalent groundwater management problem. In both applications a performance measure is either minimized or maximized by adjusting certain input parameters or design variables. For example, model calibration consists of reducing the differences between the simulation results and the measurements, such as water potentials or temperatures, by adjusting the model input parameters, such as the absolute permeability or thermal conductivity of the porous medium. A cleanup operation can be improved by minimizing, for example, the remediation time, which can be achieved by increasing pumping rates or steam temperature. Increasing pumping rates or steam temperature, however, leads to higher energy costs. Therefore, the objective function to be minimized should also reflect these costs in order to obtain an optimal remediation design.

Solving an optimization problem occurs in two steps. First, a performance measure has to be defined which is an arbitrary function of T2VOC output variables, and which depends on certain T2VOC input parameters. This objective function contains contributions from different sources which have to be appropriately weighted against each other. Secondly, a minimization algorithm is needed which is capable of updating T2VOC input parameters in order to reduce the value of the objective function. Note that the objective function is usually highly non-linear, and - in certain cases - even discontinuous. In the remainder of this section we present the general form of the objective function and discuss the bases of the minimization algorithm.

We first introduce the residual vector **r** which holds the differences between the measured and calculated state variables in the case of model calibration, and the differences between the desired and calculated costs in the case of design optimization:

$$\mathbf{r} = \mathbf{y}^* - \mathbf{y}(\mathbf{p}) \quad . \tag{11}$$

In the case of model calibration, the vector $\mathbf{y}^*$ contains the actual data. For test design, the elements of vector $\mathbf{y}^*$ are synthetically generated data, and for management problems, they represent target quantities such as zero costs or the maximum possible amount of contaminant removed from the aquifer. Note that $\mathbf{y}(\mathbf{p})$ is a function of the input parameters which are summarized in vector **p**. The elements of the parameter vector **p** are either hydrogeologic properties, initial and boundary conditions, or pumping and injection rates. In general the residuals have different units and may cover many orders of magnitudes. This requires specifying a weighting matrix of dimensions $m \times m$, where m is the total number of residuals. In inverse modeling, the weighting matrix **W** can be considered to be the inverse of the covariance matrix **C** holding measurement errors:

$$\mathbf{W} = \mathbf{C}^{-1} \quad . \tag{12}$$

If no correlations are present, the vector of the weighted residuals **z** contains elements of the form

$$z_i = \frac{r_i^* - r_i}{c_i} \quad , \tag{13}$$

where c_i is the square root of the i-th diagonal element of matrix **C**. The objective function to be minimized can be defined as follows:

$$Z = \sum_{i=1}^{m} \rho(z_i) \quad . \tag{14}$$

In maximum likelihood theory, the function ρ reflects the negative logarithm of the probability density function of z. For normally distributed residuals, we obtain

$$\rho_i = x_i^{\,2} \tag{15}$$

which yields the standard least squares estimator. The so-called L_1-estimator is obtained by minimizing the sum of the absolute values of the weighted residuals, thus

$$\rho_i = |x_i| \quad . \tag{16}$$

The choice of an appropriate estimator is somewhat arbitrary. For inverse modeling applications, it is common to use least squares or one of the robust estimators described in [9]. For management problems, i.e. if r_i represents a cost function, the L_1-estimator is most frequently applied.

Next an algorithm has to be developed that minimizes the objective function Z. Note that the functions $\mathbf{y}(\mathbf{p})$ are highly nonlinear which immediately renders the direct solution of the minimization problem infeasible. The iterative procedure involves computing a correction vector $\Delta\mathbf{p}_k$ such that the new parameter set

$$\mathbf{p}_{k+1} = \mathbf{p}_k + \Delta\mathbf{p}_k \tag{17}$$

leads to a reduced objective function, $Z_{k+1} < Z_k$, at each iteration k. A number of minimization algorithms are available to solve non-linear least-squares problems as defined by Eqs. (14) and (15) [10]. One of the most general and most robust methods is the Levenberg-Marquardt modification of the Gauss-Newton minimization algorithm [11, 12]. This procedure involves solving the following system of equations:

$$(\mathbf{J}_k^{T}\,\mathbf{W}\,\mathbf{J}_k + \lambda\cdot\mathbf{D}_k)\cdot\Delta\mathbf{p}_k = -\,\mathbf{J}_k^{T}\,\mathbf{W}\,\mathbf{r}_k \quad . \tag{18}$$

Here, $\mathbf{J}$ is the sensitivity matrix with elements $J_{ij} = \partial y_i/\partial p_j$. $\mathbf{D}$ denotes a matrix of order n (n being the number of parameters to be estimated) with elements equivalent to the diagonal elements of matrix ($\mathbf{J}_k^T\,\mathbf{W}\,\mathbf{J}_k$). The basic idea of this method is to move in the parameter space along the steepest descent direction far from the minimum, and switching continuously to the Gauss-Newton algorithm as the minimum is approached. This is achieved by decreasing the scalar λ, known as the Levenberg parameter, if an iteration is successful and increasing it if the objective function is increased. The minimum of the objective function, which may be only a local one, is detected iteratively by solving Eq. (18) for $\Delta\mathbf{p}_k$. The parameter set is then updated according to Eq. (17). Under the assumption of normality and linearity, a detailed error analysis of the final residuals and the estimated parameters can be conducted (for details see [13]).

Note that the Levenberg-Marquardt algorithm is also general enough to solve management problems usually formulated in terms of the L_1-estimator, i.e. using Eqs. (14) and (16). However, there are two shortcomings that may prevent one from using the Levenberg-Marquardt method for arbitrary cost functions. First, in the presence of multiple local minima it cannot be assured that the optimum solution is detected. While this difficulty can be alleviated, the uncertainty in the definition of the cost function and the qualitative nature of the design process in general does not make it absolutely necessary to identify the global minimum. The second problem, however, precludes the use of the Levenberg-Marquardt algorithm for certain types of cost functions. The calculation of matrix $\mathbf{J}$ requires that the functions $\mathbf{y}(\mathbf{p})$ are continuous. In remediation technology, the cost functions are often discontinuous or even discrete. For example, installation costs depend on the number of wells being drilled, making the total cost a discontinuous function if the number of wells is subjected to the optimization process. In these cases, a minimization method is required that does not rely on the calculation of derivatives. The method of simulated annealing is able to handle discontinuous objective functions that exhibit many local minima. The term simulated annealing is used by analogy to thermodynamics, specifically with the way metals slowly cool and anneal. The analogy and algorithm is described in [14]. The basic idea is to take a random step $\Delta\mathbf{p}$ which is always accepted if it leads to a reduction of the objective function. Furthermore, an uphill move is sometimes accepted with a certain, decreasing probability enabling the algorithm to escape from local minima. There is no need to calculate derivatives which makes simulated annealing applicable to several types of objective functions (discrete, discontinuous, or continuous). The main drawback of simulated annealing is its inefficiency, a result of the randomness of step $\Delta\mathbf{p}$ which almost always proposes an uphill move.

The minimization algorithms discussed in this section have been implemented in a computer program named ITOUGH2 [1]. ITOUGH2 performs automatic model calibration for the TOUGH2 family of codes [2], including T2VOC [1]. Furthermore, ITOUGH2 supports sensitivity analysis, the design of experiments, and provides uncertainties for model predictions using linear error propagation analysis or Monte Carlo simulations. It can also be used to solve groundwater management problems such as the design of a cleanup operation [15]. In the remainder of this paper we demonstrate that the combination of sophisticated process simulation and efficient and robust optimization techniques is a powerful tool to study subsurface flow and transport problems.

MODELING STUDY

The illustrative sample problem discussed in this modeling study deals with contaminant migration from a dense solvent trichloroethylene (TCE) spill into the unsaturated zone of a highly heterogeneous soil, followed by a remediation operation using air sparging and soil vapor extraction. The effect of subsurface barriers on cleanup efficiency is also studied.

We consider a shallow heterogeneous deposit containing both a saturated and a unsaturated zone. The water table is at depth of 12 m and exhibits a head gradient of about 0.03. The model domain has dimensions of 30 m × 30 m × 20 m in the x, y, and z directions, respectively, and is discretized in 15 × 15 × 20 = 4500 grid blocks. A spatially correlated permeability field is generated using simulated annealing techniques. A log-normal permeability distribution is assumed with a geometric mean of 10^{-10} m^2 and a standard deviation of one order of magnitude. During the annealing process, the permeability field is rearranged in a random manner, and the difference between the resulting variogram and a prescribed variogram is minimized using the algorithm described in the previous section. We prescribed a spherical variogram with a correlation length of 20 m in the horizontal direction. No vertical correlation is considered. The generated permeabilities were then ordered into 9 groups, covering values from 10^{-12} to 10^{-8} m^2. Porosities are in the range from 0.27 to 0.43, and are assumed to vary in proportion to the logarithm of permeability. The capillary pressure was scaled in inverse proportion to the logarithm of permeability [16]. The permeability distribution is shown in Figure 1.

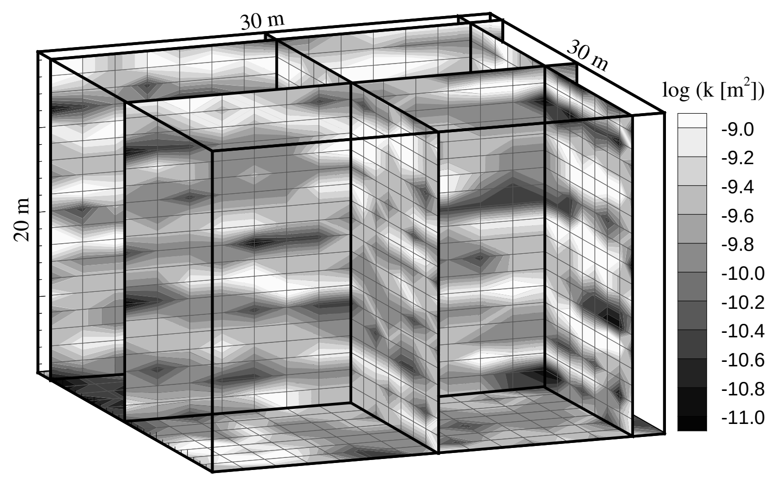

Fig. 1 Permeability distribution

The first part of the simulation aims at developing a gravity-capillary equilibrium in a saturated-unsaturated flow system that exhibits a pressure gradient in the saturated zone. An atmospheric boundary condition is applied at the surface, and a single-phase liquid Dirichlet boundary condition with a hydrostatic pressure distribution is specified at the lower part of the two opposite faces of the model. The depth to the water table is fixed at -12 m at the back face, and at -13 m at the front face. The left and right vertical faces as well as the bottom are modeled as no flow boundaries. After reaching steady-state flow conditions, the simulation continues by spilling a total of 6000 kg of TCE at the center of the model domain, 2 m below ground surface, for a period of 2 weeks.

The next part of the simulation allows for free redistribution of TCE over a 6 week period under the combined action of gravitational, capillary and pressure forces. The NAPL phase slowly slumps downward and also spreads due to capillary forces, leading to somewhat reduced NAPL contents compared to the situation immediately after the spill. Part of the NAPL plume evaporates, and contaminated soil gas of higher density flows downwards until it reaches the water table where it spreads horizontally. This density effect prevents the contaminant from being discharged naturally to the atmosphere. High aqueous TCE contents are observed near the water table, and the contaminated groundwater flows downstream and deeper into the saturated zone due to increased water density. Figure 2 shows the NAPL content (the product of NAPL saturation and porosity), TCE content in the gas phase (the product of VOC concentration in the gas phase [kg/m^3], gas saturation, and porosity), and the TCE content in the aqueous phase (the product of TCE concentration in the aqueous phase [kg/m^3], water saturation, and porosity) after the redistribution period. Note that the concentration of TCE in one phase is related to the concentration in another phase due to an assumption of local chemical equilibria. TCE vapors spreading into the system lead to a contamination of the pore water due to the solubility of the chemical in water. At this point in time, a preliminary cleanup operation is designed consisting of air sparging in combination with soil vapor extraction. In addition, four vertical cutoff walls are emplaced to contain the contaminant plume.

In the next step of the process, a cost function is defined which includes pumping costs for the air sparging and soil vapor extraction system, energy costs for the heating system, and treatment costs for the contaminated fluid produced at the pumping well. Note that all these costs depend on the flow and transport behavior in the subsurface which determines the time required to reach cleanup standards. Simulated pressures and concentrations at the injection and production well also affect the cost function. Minimization of the cost function is accomplished by determining three operational input parameters, namely the injection rate and temperature of the air sparging system, and the pumping rate at the extraction well. The results of the optimization process are summarized in Table 2. Increasing both the injection and extraction rate leads to a shorter cleanup time which reduces overall costs, despite the fact that the specific pumping and energy costs are higher. In this example, injecting air at high temperatures is not effective enough to compensate for the increased energy costs. Consequently, ITOUGH2 proposes to use air at ambient temperatures. Overall the proposed design is about twice as efficient as the original design.

Table 2 Set of proposed operational parameters determined by minimizing the cost function

Parameter	Original Design	Optimized Design
Injection rate [kg/s]	0.01	0.03
Injection temperature [˚C]	90.00	40.00
Extraction rate [kg/s]	0.01	0.04
Cost function ["$"]	1090.00	550.00

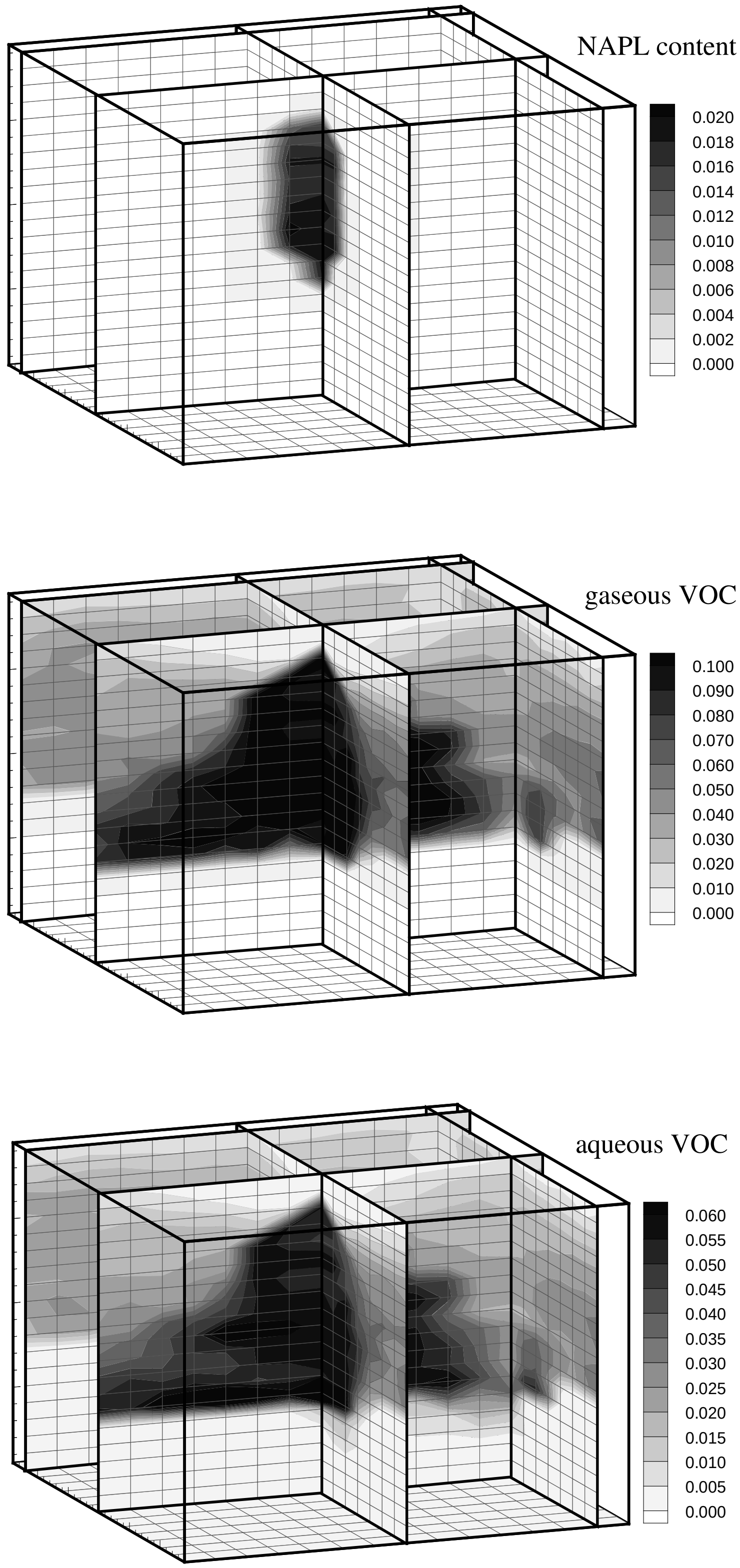

Fig. 3 NAPL content (top), vaporized VOC content (center), and dissolved VOC content (bottom) after redistribution

CONCLUDING REMARKS

A series of computer codes has been developed to support the design of a multi-component cleanup system. Accurate simulation capabilities for flow and transport of contaminants under multiphase flow conditions are crucial for predicting the fate of the contaminant for a given remediation scenario. Inverse modeling techniques provide an essential tool for site characterization by determining model-related input parameters for the numerical simulator. The same optimization algorithms can also be used to study alternative cleanup operations by minimizing a cost function that reflects the efficiency of the proposed design. The results of such a simulation-optimization study can be used as a basis for the evaluation and optimization of an integrated remediation technology.

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