A robust approach for iterative contaminant source location and release history recovery

Alexander Y. Sun *, Scott L. Painter, Gordon W. Wittmeyer

Geosciences and Engineering Division, Southwest Research Institute®, 6220 Culebra Road, San Antonio, TX 78238, United States

Received 9 January 2006; received in revised form 8 June 2006; accepted 20 June 2006
Available online 4 August 2006

Abstract

Contamination source identification is a crucial step in environmental remediation. The exact contaminant source locations and release histories are often unknown due to lack of records and therefore must be identified through inversion. Coupled source location and release history identification is a complex nonlinear optimization problem. Existing strategies for contaminant source identification have important practical limitations. In many studies, analytical solutions for point sources are used; the problem is often formulated and solved via nonlinear optimization; and model uncertainty is seldom considered. In practice, model uncertainty can be significant because of the uncertainty in model structure and parameters, and the error in numerical solutions. An inaccurate model can lead to erroneous inversion of contaminant sources. In this work, a constrained robust least squares (CRLS) estimator is combined with a branch-and-bound global optimization solver for iteratively identifying source release histories and source locations. CRLS is used for source release history recovery and the global optimization solver is used for location search. CRLS is a robust estimator that was developed to incorporate directly a modeler’s prior knowledge of model uncertainty and measurement error. The robustness of CRLS is essential for systems that are ill-conditioned. Because of this decoupling, the total solution time can be reduced significantly. Our numerical experiments show that the combination of CRLS with the global optimization solver achieved better performance than the combination of a non-robust estimator, i.e., the nonnegative least squares (NNLS) method, with the same solver.

© 2006 Elsevier B.V. All rights reserved.

Keywords: Contaminant source identification; Model uncertainty; Constrained robust least squares; Robust programming; Global optimization; Branch and bound
1. Introduction

Remediation of contaminated aquifers is costly and time-consuming. The effectiveness of any remediation plan relies heavily on knowledge of contamination sources. Only after contaminant sources are identified can (i) remediation costs be partitioned among liable parties, (ii) the most appropriate remediation technology be chosen, and (iii) the contaminant transport pathways be identified. The need for contaminant source identification arises when site historic records alone are insufficient to recover the sources. Source identification is further complicated because contaminants are usually not detected until long after they are released into aquifers, at which time the contaminant plume may have already migrated a significant distance from the source. Because of these complications, certain inversion technique must be used to reconstruct source locations and release histories from site hydrogeologic models and concentration measurements.

Like many other problems, contaminant source identification is inherently ill-posed, meaning a small perturbation in the input may result an arbitrary large perturbation in the solution (Sun, 1994; Sun and Sun, 2005). Discretization of ill-posed inverse problems inevitably leads to ill-conditioned systems that are very sensitive to perturbations. Perturbations in contaminant source identification can be caused by limitations in data quantity and quality, and by model uncertainty, which may be non-Gaussian and systematic. Common model uncertainties include parameter uncertainty and model structure uncertainty. The former may be caused by the variability of hydrogeologic parameters manifested at different spatial scales, whereas the latter is due to incomplete knowledge or incorrect conceptualization of system features, events, and processes (Meyer et al., 2004). Errors incurred by numerical models pose another source of uncertainty.

Various deterministic and statistical inversion algorithms have been proposed in the last two decades for solving source identification problems. Recent surveys of approaches can be found in Atmadja and Bagtzoglou (2001) and Michalak and Kitanidis (2004). Many of the previous studies assume that source locations are known, and thus, only focus on developing a special technique for identifying the initial concentration distributions or contaminant release histories. Examples of such techniques include the least squares (LS) approach (e.g., Gorelick et al., 1983); the Tikhonov regularization (TR) approach (e.g., Skaggs and Kabala, 1994, 1998), the minimum relative entropy approach (e.g., Woodbury and Ulrych, 1996; Woodbury et al., 1998), and the geostatistical inversion approach (Snodgrass and Kitanidis, 1997; Michalak and Kitanidis, 2004). The backward probabilistic model (Neupauer and Wilson, 1999, 2001) yields backward location probability distributions based on concentration measurements obtained at monitoring wells. It, however, is restricted to a single unknown point source. Mahar and Datta (2001) used an embedded model approach to identify both flow model parameters and source terms by solving a nonlinear optimization problem. The computational cost associated with such a technique can be enormous.

Most techniques mentioned above assume that the underlying model is known perfectly. The effects of model uncertainty are mainly addressed through ad hoc sensitivity analyses. Sensitivity analysis quantifies locally the stability of the nominal solution with respect to infinitesimal data perturbations, but it does not address stability improvement (Bertsimas and Sim, 2004). As an alternative to sensitivity analysis, we incorporate the a priori information on model uncertainty directly into estimation. Specifically, we examine cases in which the model perturbation is unknown, but is deterministically bounded. Parameter estimation under bounded uncertainty has already been applied to, for example, engineering structural analysis (Ben-Tal and Nemirovski, 1997), portfolio optimization (Bertsimas and Sim, 2004), and robust controller design (El Ghaoui and Lebret, 1997; De Fonseca et al., 2001). In general, these methods seek to minimize the worst-case residual across all bounded perturbations of a model by solving a minimax problem.
Sun et al. (2006a) recently developed a robust framework for contaminant source identification. The contaminant source identification problem was first cast into one of solving linear equations, where the response matrix was constructed using a superposition technique. This formulation is general and can be implemented using any porous media flow and transport solvers. A CRLS estimator was then formulated to obtain estimation under uncertainty. As its name implies, CRLS is a deterministic, robust estimator. Robust estimation seeks a solution that remains feasible with high probability even when some parameters deviate from their nominal values.

The main features of CRLS are: (i) model uncertainty is included as part of the analysis, (ii) the bound of model uncertainty is used to rigorously determine a regularization parameter that controls the size of allowable corrections to the nominal system, and (iii) the solution is constrained by both second-order cone constraints and linear constraints. The Monte Carlo simulations conducted by Sun et al. (2006a) showed that CRLS gave better performance than its non-robust counterpart, i.e., the NNLS estimator, for ill-conditioned systems.

Oftentimes, source locations are unknown as well. Although in principle, source location identification can be solved by including all possible source locations as legitimate guesses and then eliminating those locations that show null release histories, the search strategy suffers from a number of drawbacks: first, it requires a significant amount of prior knowledge of potential source locations; and second, it is neither automatic nor optimal. A better, although more computationally demanding, approach is to include source characteristics (e.g., location and shape geometry) as decision variables. The resulting optimization problem is nonlinear and nonconvex, which often necessitates using a global optimization solver to prevent the objective function from being trapped at local minima. Aral et al. (2001) considered coupled identification of point source locations and release histories using a progressive genetic algorithm (GA). Their approach essentially transforms a nonlinear optimization problem into a number of quadratic optimization problems with linear constraints, which are subsequently solved by GA. A GA-based method generally maintains a population of points for every generation and uses genetic operators, such as crossovers and mutations, to generate new points. These old and new points compete for survival in the next generation based on a fitness function. In principle, GA examines every point from every other point in a search space. A drawback is that it may take a long time for GA to converge once it has reached the neighborhood of a global optimum. Aral et al. (2001) illustrated their approach with a single source example. They found that including source locations as decision variables significantly affects source identification due to the interaction between the release at a variable source location and the observation data. Mahinthakumar and Sayeed (2005) addressed the source identification problem using GA combined with a local search algorithm. Using this approach, the size, location, and initial concentration of a single source were identified simultaneously. A local search was used to fine-tune the GA solution and code parallelization was implemented because the computation time of a serial solver was too long to be acceptable.

The effect of model uncertainty was precluded in most previous studies on source location identification. In this project, we combine CRLS with a global optimization solver to iteratively identify source locations and release histories. This iterative source identification approach leverages the robustness of CRLS, and it is more efficient and practical than the embedded model approach (e.g., Mahar and Datta, 2001) because the decoupling between source release history identification and location identification reduces the dimension of the search space. This reduction can be especially significant when the number of release periods is large.

This paper is organized as follows: first, the formulation of our robust framework for source release history recovery is recapitulated; second, the iterative approach for source location and release history recovery is presented; and finally, the iterative approach is demonstrated via numerical examples.
2. A robust framework for source release history recovery

For completeness, we briefly describe the formulation of our robust framework for source release history identification in this section. More details can be found in Sun et al. (2006a).

2.1. Formulation of the Unit Pulse Response Matrix (UPRM)

Consider the identification of contaminant release history from multiple locations. The first step is to parameterize the unknown release history. We adopt a form that is specifically designed for MT3DMS (Zheng and Wang, 1999).

Assume \( M \) contaminant sources are located at \( \Omega_1, \Omega_2, \ldots, \Omega_M \), respectively, and the unknown release history of these \( M \) sources during \( N \) time periods, \( T_1, T_2, \ldots, T_N \), is

\[
\mathbf{z} = \{z_{mn}\}, n = 1, 2, \ldots, N; m = 1, 2, \ldots, M
\]

(1)

where \( \mathbf{z} \in \mathbb{R}^{K \times 1} \), \( \mathbb{R} \) is the real space, \( K = M \times N \) is the total number of unknowns, and \( z_{mn} \) is the strength of the \( m \)-th source during \( n \)-th time period. A double index is used on \( \mathbf{z} \) for mathematical clarity. Throughout this work, however, \( \mathbf{z} \) is treated as a vector and is stored in row-order, i.e., \( \{z_{11}, z_{12}, \ldots, z_{1N}, z_{21}, z_{22}, \ldots, z_{2N}, \ldots, z_{M1}, z_{M2}, \ldots, z_{MN}\} \).

Mass transport in porous media can be modeled by the following advection dispersion equation (cf., Bear, 1979; Sun, 1996)

\[
\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i} \left( D_{ij} \frac{\partial C}{\partial x_j} \right) - \frac{\partial}{\partial x_i} (V_i C) + \sum_{m=1}^{M} \sum_{n=1}^{N} z_{mn} \delta(\Omega_m) \delta(T_n), i, j = 1, 2, 3
\]

(2)

with initial and boundary conditions

\[
C(x, 0) = 0, x \in \Phi
\]

\[
C(x, t) = 0, x \in \Gamma_1, 0 \leq t \leq T
\]

\[
\frac{\partial C}{\partial v} = 0, x \in \Gamma_2, 0 \leq t \leq T
\]

(3)

In Eq. (2), \( t \) is time; \( x \) is the spatial coordinate vector; \( C(x, t) ([M/L^3]) \) is the concentration distribution at \( t \); \( D_{ij} ([L^2/T]) \) are the components of the dispersion tensor, \( \mathbf{D} \); \( V_i ([L/T]) \) are the components of the seepage velocity vector, \( \mathbf{V} \); \( \delta(\cdot) \) are the Kronecker deltas; and the double summation term on the right-hand side represents parameterization of the unknown source function. In Eq. (3), \( \Phi \) denotes the flow region with boundary segments \( \Gamma_1 \) and \( \Gamma_2 \), \( v \) is the normal vector to the boundaries, and \( T \) is the total time of interest. Although the reaction term is not included in Eq. (2) for clarity, the formulation presented here is valid for mass transport involving any type of linear reactions. Identification of initial source concentration corresponding to, for example, catastrophic releases, is a special case of the more general case considered here.

From the perspective of system identification, the UPRM approach essentially links system responses to a series of unit-strength inputs. This is possible if the system is linear, which is true under the assumption of the current application. The concentration field can be expressed as

\[
C(x, t; \mathbf{z}) = \sum_{m=1}^{M} \sum_{n=1}^{N} z_{mn} C_m^0(x, t)
\]

(4)
where \( C_{mn}^0(x, t) \) is the concentration field resulting from a unit-concentration source in a single release period. Assuming there are \( L \) concentration measurements, which are written as \( C_{l}^{ob}(x_l, t_l) \) \((l = 1, \ldots, L)\), the link between system inputs and outputs is established via the following system of equations,

\[
Az \approx b
\]

where \( A \in \mathbb{R}^{L \times K} \) is the response matrix, and \( b \in \mathbb{R}^{L \times 1} \) is vector containing measurement data. In general, \( K \) model runs are needed to identify \( K \) sources. As will be shown later, a significant simplification is possible when the flow field is steady and all release periods have equal lengths.

2.2. Model uncertainty and CRLS

Methods for solving systems of linear equations like Eq. (5) have been studied for a long time. It is well known that the ordinary LS approach gives the best unbiased solutions if the data noise is independent and identically distributed. If \( A \) is ill-conditioned, certain regularization techniques, such as TR (Tikhonov and Arsenin, 1977), must be applied to stabilize the solution. A problem with the standard TR formulation is that there is no rigorous way to determine the best regularization parameter. As a result, TR may yield over- or under-regularized solutions, and thus, reduce confidence in the final result.

The need thus arises for an estimator that considers errors in both \( A \) and \( b \), and is robust for ill-conditioned systems. Note that when a system is ill-conditioned, a meaningful approximation can still be obtained for it. However, standard methods, such as ordinary LS, cannot be used in a straightforward manner for such a system (Hansen, 1994).

The robust least squares (RLS) method, which was first formulated concurrently by El Ghaoui and Lebret (1997) and Chandrasekaran et al. (1996), optimally regularizes the corrections to \( A \) and \( b \), using \textit{a priori} bounds on the uncertainty. The original RLS proposed by El Ghaoui and Lebret (1997) solves a minimax optimization problem,

\[
\min_z \max_{||\Delta A : \Delta b||_F \leq \rho} \|(A + \Delta A)z - (b + \Delta b)\|_2
\]

where \( \Delta A \) and \( \Delta b \) are the unknown errors (or perturbations) in \( A \) and \( b \); \( \rho \) is the upper bound of the augmented error matrix, \([\Delta A : \Delta b]\), which is formed by appending \( \Delta b \) after the last column of \( \Delta A \); and \( ||\cdot||_2 \) and \( ||\cdot||_F \) denote the matrix \( L-2 \) norm and Frobenius-norm, respectively (Golub and Van Loan, 1989). The Frobenius norm of a matrix is the square root of the sum of the squares of its elements. RLS minimizes the worst-case perturbation in the Frobenius-norm such that the effect of uncertainty will never be overestimated beyond what is reasonably assumed by the \textit{a priori} uncertainty bounds (Chandrasekaran et al., 1996).

It is not clear that putting a bound on the matrix \([\Delta A : \Delta b]\) is natural. A more meaningful way is to assume that \( \Delta A \) and \( \Delta b \) are subject to separate bounds. In other words, we are interested in solving the following minimax problem instead,

\[
\min_z \max_{\Delta A, \Delta b} \|(A + \Delta A)z - (b + \Delta b)\|_2
\]

subject to \( ||\Delta A||_2 \leq \rho_A \) and \( ||\Delta B||_2 \leq \rho_B \)
where \( \rho_A \) and \( \rho_b \) are bounds of the \( L-2 \) norms of \( \Delta A \) and \( \Delta b \), respectively. It can be shown (Sun et al., 2006a) that this minimax problem is equivalent to the following second-order cone programming (SOCP) problem,

\[
\begin{align*}
\min \eta \\
\text{subject to} \\
\|Az - b\|_2 \leq \eta - \tau \quad \text{and} \quad \rho_A \|z\|_2 \leq \tau \\
\text{and} \quad lb_k \leq z_k \leq ub_k, \quad k = 1, \ldots, K
\end{align*}
\]  

where \( \eta \) and \( \tau \) are slack variables introduced to facilitate the problem solving. The last constraint in Eq. (8) is added to enforce nonnegativity, where \( lb_k \) and \( ub_k \) are the lower- and upper-bound of the \( k \)-th component in the unknown vector, \( z \). Simply speaking, SOCP problems are nonlinear, convex optimization problems in which a linear function is minimized over the intersection of an affine set and the product of second-order cones (Lobo et al., 1998; Alizadeh and Goldfarb, 2003). SOCP problems can be solved in polynomial time by the efficient primal–dual interior-point methods (Nesterov and Nemirovski, 1995; Nesterov and Todd, 1997; Fujisawa et al., 1997). All problems in this study were solved using SeDuMi (Sturm, 1998).

The bound of uncertainty plays a central role in CRLS. For demonstration purposes, we will assume in the remainder of this discussion that the model structure is correct and the uncertainty in \( A \) is caused solely by uncertainty in the hydraulic conductivity. Other types of uncertainty can be considered, as long as the bound of uncertainty can be determined. Although many methodologies exist for propagating the input uncertainty through a system (e.g., Zhang, 2001; Helton and Davis, 2003), we are mainly concerned with the parameter combination that leads to the worst deviation from the system’s nominal state. We assume that parameter uncertainty is characterized in a set-theoretic setting, and thus, can be propagated using ellipsoidal calculus or interval analyses (Kurzhanski and Vályi, 1996; Ros et al., 2002). If a system is linear and convex, which is the case we consider, only the vertices of the parameter space need to be searched to identify the worst-case perturbation.

Sun et al. (2006a) conducted one- and two-dimensional numerical experiments to compare the performance of CRLS with several classical methods (i.e., ordinary LS, standard total least squares, and NNLS). In all examples, the release history of a single source was identified. The location of the source was assumed known \textit{a priori}. CRLS was found to perform better than other estimators considered. The confidence gained from our previous study prompted us to consider simultaneous identification of source locations and release histories.

### 3. A framework for source location and release history identification

Source locations may be identified by including all potential locations in the UPRM formulation. This approach is feasible only when sufficient \textit{a priori} information is available. In many situations, modelers know at best the bounds of a region where potential sources may be located. We are, therefore, motivated to use a global optimization solver to automate the location search process and CRLS to recover the source release history for each trial point. This iterative identification strategy not only leverages the robustness of CRLS, but also reduces the dimension of the nonlinear optimization problem, as compared to the coupled identification approach.

Nonlinear optimization problems can be solved by either local or global optimization methods. Mahinthakumar and Sayeed (2005) reviewed the merits of both methods in the context of contaminant source identification. Local optimization methods are fast and, in general, scale well
for large-sized problems. However, local optimization methods rely on local gradients of the objective function to guide the search direction, and thus, tend to converge to a local minimum, rather than a global minimum (cf. Sun, 1994; Wang, 2001; Boyd and Vandenberghe, 2004). Local optimization methods are also highly dependent on initial estimates and are sensitive to algorithm parameters. In contrast, global optimization, which can be considered a stronger version of local optimization, seeks the best solution in the parameter space (cf. Neumaier, 2004). The difference between local and global minima is a practical concern in contaminant source identification because the former may not correspond to real source locations.

Numerous deterministic and stochastic global optimization algorithms exist in the literature. Neumaier (2004) classifies the global optimization algorithms into incomplete, asymptotically complete, complete, and rigorous methods. An incomplete method uses some type of heuristic strategy for searching, but provides no safeguards against trapping at local minima; whereas the other three methods reach a global minimum with certainty and differ only in the manner of reaching convergence. GA and simulated annealing (SA) (cf. Hajek, 1988), in their original forms, are considered incomplete methods by Neumaier (2004). They have become popular in research communities because they are relatively easy to understand and implement. The convergence rate of GA and SA may be exceedingly slow, and thus, they are more suitable for applications where function evaluations are not expensive. Various ad hoc enhancements have been proposed for both methods (cf. Wang, 2001). Other than GA and SA, stochastic methods based on multiple-random-start are also considered incomplete methods. These methods choose random points in the search space and perform local optimization on these points. A problem with these methods is that they may spend time sampling infeasible regions and converge to deep local minima. Branch-and-bound algorithms constitute an important class of the complete methods. A branch-and-bound method splits a problem recursively into subproblems, which may then be eliminated by showing that the subproblem cannot lead to a better solution. This can be easily verified if the lower bound of the objective function is known. Otherwise, a stopping rule can be put in place.

In-depth review of global algorithms can be found in Pintér (1996), Wang (2001), and Neumaier (2004). One should keep in mind that global optimization is not a panacea because it comes at the price of efficiency — the complexity of global optimization grows exponentially with problem size. The hope is that with a particular global optimization algorithm, one can solve a problem much faster. Any rational measure or assumption that can lead to a reduction in search space is worth exploring.

Global optimization has been applied in hydrogeology problems other than source identification. For example, Huang and Mayer (1997) investigated optimal groundwater remediation management in homogeneous and heterogeneous aquifers, using continuous pumping rates and discrete well locations as decision variables. In Huang and Mayer’s work, GA was used to search for the optimal design, and the resulting computational time was excessive. Guan and Aral (1999) identified optimal pumping well locations and pumping rates for a remediation plan using progressive GA. Maskey et al. (2002) compared the efficiency and accuracy of several global optimization algorithms for finding the optimal remediation strategy.

The constrained nonlinear optimization problem that we want to solve is constrained by the so-called box constraints,

$$\min f(\Psi)$$
subject to $\Psi \in [l_b, u_b]$
where $\Psi$ is a vector of decision variables, and the box constraints contain the lower- and upper-bound of each decision variable,

$$[l_b, u_b] = \{\Psi \in \mathbb{R}^{D_S}|l_i \leq \psi_i \leq u_i, i = 1, \ldots, D_S\}$$

in which $D_S$ is the dimension of the search space, and $l_b$ and $u_b$ are the lower- and upper-bound vectors, respectively. The decision variables in this case are parameters that uniquely define the geometry of contaminant sources. For example, if the source area is rectangular, a possible set of decision variables would be the width, length, and coordinates of the upper-left corner of the area.

A workflow diagram illustrating the iterative procedure for source location and release history identification is shown in Fig. 1. For each trial location that is selected by the location search routine, the forward problem is solved to generate the response matrix, and the release history is identified. With the identified release history, the model outputs at the observation locations can be obtained, from which the value of the objective function can be calculated. The objective function is defined in the following form,

$$f = \sum_{i=1}^{nw} [C_{i}^{\text{obs}} - C_{i}^{\text{cal}}(x_s)]^2$$

where $i$ is observation index, $nw$ is the total number of observations, $C_{i}^{\text{obs}}$ is the observed concentration, and $C_{i}^{\text{cal}}$ is the calculated concentration at an observation location and is also a function of the source location $x_s$. The stopping criteria for the search process are algorithm
dependent. In general, the search process stops when no improvement in the solution can be found after a number of successive runs, or when the maximum number of function evaluations has been exceeded.

We used an optimization package, SNOBFIT (Huyer and Neumaier, submitted for publication), to solve Eq. (9). SNOBFIT implements a branch-and-bound algorithm, and it is specifically designed for problems involving expensive function evaluations. SNOBFIT performs global searches by branching, and local searches by local quadratic fits. The algorithm generates a

Fig. 2. (a) Configuration of the synthetic problem. The locations of sources $S_1$ and $S_2$ are labeled in the figure. The search range for Example 1 is marked by the solid-line rectangle, and the range for Example 2 is marked by the dashed-line rectangle. The locations of 11 observation wells are shown with filled circles. Also shown is the steady-state head contour. (b) Observation times of all observation wells.
number of evaluation points in each step and proceeds by successively partitioning the parameter space and building local quadratic models. The search process is terminated if no better solution can be found after a specified number of successive steps, or when a given minimal objective function value is reached.

4. Numerical examples

The iterative procedure for source location and release history recovery are demonstrated through several synthetic examples. Contaminant transport in a two-dimensional, unconfined aquifer is considered, a problem that was originally used in Sun et al. (2006a) to demonstrate release history identification. Fig. 2(a) shows the problem setting, where the locations of 2 true sources and 11 observation wells are shown. Constant heads of 360 m and 60 m are applied to the upper-left and lower-right corners of the aquifer, respectively. To simulate data gaps, not all wells are sampled at all observation times. The sampling times of wells are shown in Fig. 2(b). All other model parameters are listed in Table 1. For illustration purposes, we assume that the aquifer is homogeneous and isotropic. A more complex hydraulic conductivity distribution was considered recently in Sun et al. (2006b). It is emphasized here that CRLS only requires the bound of perturbations to operate. The key is to be able to find the parameter combination that can result in the worst-case perturbation from the nominal state. We assume here that the nominal hydraulic conductivity value ($K_c$) is 25 m/day, and the actual value of $K_c$ lies in the range between 22.5 m/day and 27.5 m/day. Sun et al. (2006a) found that the maximum deviation from the nominal state always corresponds to the lower bound of $K_c$ for such a problem. The bound of $\Delta A$, $\rho_A$, can then be evaluated from the $L_2$ matrix norm, $\| A_{K_c=25} - A_{K_c=22.5} \|_2$, where $A_{K_c=25}$ and $A_{K_c=22.5}$ are the response matrices corresponding to $K_c = 25$ and 22.5 m/day, respectively. The number of model calls required is $2N + 1$ per trial location if there are $N$ release periods to be identified. The process can be simplified significantly if the flow field is steady and all release periods have equal durations. Under these conditions, only one model run is required to obtain the system response for the first period, and the system responses to all other release periods can be obtained through time shifting (cf. Aral et al., 2001). Consequently, the number of model runs per trial location is reduced to 3 for CRLS. To simplify the problem further, we assume that the source area is known and is equivalent to the size of a $2 \times 2$ grid area. In other words, only the upper-left corner of the source needs to be identified in the location search part of the problem. Based on our numerical experiments, the number of evaluation points used by SNOBFIT in each branch-and-bound step is set to $d + 4$, where $d$ is the number of unknowns; and the number of successive steps without improvement is set to 10, after which the location search is terminated.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grid dimension</td>
<td>$100 \times 50$</td>
</tr>
<tr>
<td>Cell size</td>
<td>$50 \text{ m} \times 50 \text{ m}$</td>
</tr>
<tr>
<td>Porosity</td>
<td>0.25</td>
</tr>
<tr>
<td>Longitudinal dispersivity</td>
<td>10 m</td>
</tr>
<tr>
<td>Transverse dispersivity</td>
<td>1 m</td>
</tr>
<tr>
<td>Upper and lower boundary heads</td>
<td>360 m and 60 m</td>
</tr>
<tr>
<td>Nominal conductivity</td>
<td>25 m/day</td>
</tr>
</tbody>
</table>
4.1. Identification of a single source location

The location and release history of a single source are identified in this problem. The cell indices of the upper-left corner of the true source, which is labeled as $S_1$ in Fig. 2(a), are (16, 9); the total release time of the true source is discretized into 12 equal-length periods. For demonstration, we assume that the release time structure is known. This, however, does not limit our methodology in any way because it is trivial to consider any other type of partition schemes. Although all systems considered in this example are over-determined, CRLS can be used to solve under-determined systems. An example of solving under-determined system with CRLS was given in Sun et al. (2006a, Section 4.1.3). Because of the sub-optimal sampling scheme, the resulting nominal response matrix is ill-conditioned, with a matrix condition number of $6.9 \times 10^{11}$. Before solving the problem, it is instructive to study the shape of the objective function, Eq. (11), which is plotted in Fig. 3 for the case of no measurement error. It can be observed from the figure that the global minimum is surrounded by diamond-shaped contours. In this case, the value of the objective function is very sensitive to location changes, with its range spanning more than 3 orders of magnitude in the search area.

Two numerical tests were performed. In the first test, only model uncertainty exists, which is assumed to be caused by the parameter uncertainty in $K_c$. In the second test, the measured concentrations are also subject to a maximum of 10% random relative measurement error, in addition to the model uncertainty. The lower- and upper-bounds of the search range, which is marked by the rectangular region inside the solid line in Fig. 2(a), were set to $\mathbf{l}_b = [10, 5]$ and $\mathbf{u}_b =$
[20, 15] in both tests. We used the following formula to calculate the total estimation error (Mahinthakumar and Sayeed, 2005),

\[
\frac{1}{np} \sum_{i}^{np} \left| p_i - p_i^e \right| \times 100\%
\]  

(12)

where \(p_i\) and \(p_i^e\) are the true and estimated decision variables, and \(np\) is the total number of unknowns. Eq. (12) has the advantage of not biasing toward parameters with large values, but it may be inappropriate when the estimates are close to zero.

In the first test, both the location and release history of the true source were recovered almost exactly after 64 trial locations. In the second test, the true source location was found after 62 trial locations, but the total estimation error was 7.5%, which was caused by errors in the identified release history. For comparison, we replaced CRLS with NNLS and ran the second test again. Because NNLS does not provide any mechanism for addressing parameter uncertainty, in principle any \(K_c\) value within the specified bound is possible. For the same set of measurements and a \(K_c\) value of 23 m/day, SNOBFIT (now coupled with NNLS) fully recovered the source location after 61 trial locations. The estimation error was 50.5% and can be attributed entirely to misfits in the identified release history. The identified release histories from the two methods are plotted in Fig. 4, together with the true release history. Although only one set of random measurement errors was used in the tests, we expect the superiority of CRLS over NNLS to stay for other realizations of random errors, an observation that has been verified in Sun et al. (2006a) through Monte Carlo simulation.

![Fig. 4. Comparison between the true release history (solid line), and the release histories identified by CRLS (dotted line with ×) and NNLS (dashed line with ○) for the single-source location example. The release history is divided into 12 release periods.](image)
In short summary, the iterative search procedure was able to recover the single source location exactly, even in the presence of measurement error, because the objective function was relatively sensitive to source location changes. More importantly, a deviation from the true source location caused a monotonic increase in the objective function, which in turn helped the location search process. The instability of NNLS was not severe enough to interfere with the location search process in this example.

4.2. Identification of two source locations

The location of the single source was recovered accurately in the previous example. A question is whether this remains true for multiple source locations. To test this, we added another source, labeled as $S_2$ in Fig. 2(b), to the previous problem. The upper-left corner of the newly added source is located at cell indices (22, 12). The two sources have the same release history structures, but different strengths over time. The condition number of the resulting nominal response matrix corresponding to this case is $3.23 \times 10^5$. The lower- and upper-bounds of the search ranges for the four cell indices, i.e., ($x_1$, $y_1$, $x_2$, $y_2$), were set to $l_b = [11, 6, 11, 6]$ and $u_b = [28, 16, 28, 16]$, respectively (see the rectangular region surrounded by the dashed line in Fig. 2a).

When no measurement error was present, the combination of SNOBFIT and CRLS recovered the true locations after sampling 110 different locations. The estimation error, which resulted from misfits in the identified release history, was 7.5%. When the same test was run using NNLS for a $K_c$ value of 23 m/day, the identified source locations were (22, 10, 21, 12) and the total estimation error was 69%. In this case, the instability of NNLS became severe enough to interfere with the

![Figure 5](image)

Fig. 5. Comparison between the true release history (solid line) and the release histories identified by CRLS (dotted line with ×) and NNLS (dashed line with ○) in the two source location example. The upper plot is for the source located at cell numbers (16, 9) and the lower plot is for the source located at (22, 12). The total number of release periods is 12 and the measurements are assumed exact.
location search process. In contrast, the robustness of CRLS enabled us to find the true source locations. The recovered release histories by both methods are shown in Fig. 5.

When a 10% random error was added to the measured concentrations, the combination of SNOBFIT with CRLS converged to locations (15, 9, 21, 12), and the total estimation error was 30%. Interestingly, with NNLS, the identified locations were (15, 9, 23, 12), and the total estimation error was 43%, which is lower than in the case of zero measurement error. The recovered source release histories by the two methods are shown in Fig. 6. One possible explanation for the apparent improvement of NNLS in this case is that the specific set of random numbers used offset the instability of NNLS in some degree. Indeed, additional experiments performed with different sets of random numbers validated this hypothesis — the estimation error of CRLS was little changed, while the estimation error of NNLS fluctuated tremendously.

Some conclusions that can be drawn from this experiment are: (i) the optimization problem became considerably more difficult even with one extra source; (ii) it is unreliable to couple SNOBFIT with a non-robust estimator. The running time of most numerical tests on a computer equipped with Intel Centrino 1.5 MHz CPU was under 3 h. MODFLOW 2000 (Harbaugh et al., 2000) was used to solve the flow problem and MT3DMS (Zheng and Wang, 1999) was used to solve the transport problem.

5. Summary

Coupled source location and release history recovery is a nonlinear, nonconvex optimization problem that can be difficult to solve for high-dimension parameter space. In addition, source identification is adversely affected by model uncertainty, which can be significant. This work presents an approach for iteratively recovering contaminant source locations and release histories.
The approach combines a global search algorithm with a robust estimator, CRLS, for iterative source location and release history recovery. CRLS assumes that both measurement error and model uncertainty exist, and it is robust against the worst-case perturbation. One of the main advantages of the iterative approach is that the dimension of the global search space is dramatically reduced, especially when the number of release periods is large. This is crucial for global search routines because they usually converge slowly in high-dimensional search space. The robustness offered by CRLS has been shown to counteract the instability caused by model uncertainty and source location changes, and thus, to facilitate the recovery of source locations and release histories.

Acknowledgment

Funding for this project is provided by the Advisory Committee for Research at Southwest Research Institute® (01.20.R9530). The authors want to thank our colleague, Dr. Osvaldo Pensado, the Editor-in-Charge, and two anonymous reviewers, for their constructive comments.

References