NEAR REAL-TIME ATMOSPHERIC CONTAMINATION SOURCE IDENTIFICATION BY AN OPTIMIZATION-BASED INVERSE METHOD

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ABSTRACT

In this paper we propose a method to identify contamination events (location and time of release) by enhancing a mathematical method originally proposed by Carasso *et al.* (1978). The method of the *Marching-Jury Backward Beam/Plate Equation*, which was previously applied to groundwater problems (Atmadja and Bagtzoglou, 2001a; Bagtzoglou and Atmadja, 2003; Cornacchiulo and Bagtzoglou, 2002) is enhanced and coupled to discrete Fourier transform processing techniques to solve a two-dimensional (2D) advection-dispersion transport problem with homogeneous and isotropic parameters backwards in time. The difficulties associated with this ill-posed, inverse problem are well recognized (Atmadja and Bagtzoglou, 2001b).

We, therefore, enhance the method by integrating an optimization scheme that takes as input parameters the stabilization parameter and the coefficient of diffusion. The objective function is set as an equally weighted sum of different mass and peak errors that can be calculated based on a combination of exhaustive contaminant coverage at specific points in time (e.g., lidar) and/or point data collected at a continuously monitored network of chemical sensors or biosensors, which may be stationary or mobile.

INTRODUCTION

In the aftermath of the catastrophic events of September 11, 2001 and the international anthrax and sarin nerve gas scare, it has become clear that no reliable method currently exists to protect ordinary citizens from chemical and biological agents that are dispersed in the air. The challenge involves both the detection and identification of possible contamination sources in the shortest time possible. Therefore, a goal of paramount importance for the international security agencies is to infer in near real-time the existence of possible threats and subsequently track and intercept potential targets. Sensor systems, capable of simultaneously monitoring the concentrations of multiple, related air-borne toxins have been and are continuously being developed. However, methods that are capable of utilizing the sensor information in real-time for identifying the source and time of release for a contamination event do not exist.

The primary goal of this research effort is to develop a method for identifying as quickly as possible pollution sources that could be the result of terrorist attacks in any component of the hydrologic cycle (i.e., watersheds, groundwater, reservoirs), but it is expected that it could also be applied to any other diffusion-like problems such as atmospheric air pollution. This type of problem has long been recognized for being extremely challenging since the solution relies on solving the governing equation for diffusion backwards in time, which belongs to the category of ill-posed problems (Atmadja and Bagtzoglou, 2001b).

In this paper, we study the 2D advection-dispersion transport problem of a plume of unit mass that was instantaneously released at an unknown location (ξ, η). The well-known general equation for such a problem is described by:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left[D_x \frac{\partial C}{\partial x}\right] + \frac{\partial}{\partial y}\left[D_y \frac{\partial C}{\partial y}\right] - \frac{\partial}{\partial x}[uC] - \frac{\partial}{\partial y}[vC] \tag{1}$$

subject to appropriate initial and boundary conditions. The parameter of interest C represents the solute concentration and the terms D_x, D_y and u, v are the dispersion coefficients and transport velocity components in the x and y directions, which are further assumed to be aligned with the principal axes of the dispersion tensor, respectively.

We then study the performance of our algorithm using a typical analytical solution for a 2D problem based on a Gaussian dispersion model with homogeneous and isotropic coefficients. More specifically, we present the results of two different scenarios for which the velocity term is known *a priori* in both directions. In the first case the observation or conditioning point follows the center of mass of the plume. This case is analogous to a mobile environmental laboratory tracking the plume while transmitting concentration information. The second case entails a set of observation points that are fixed in space and is analogous to the plume being tracked by a network of bio- or chemical sensors that is fixed in space. We apply the *Marching-Jury Backward Beam or Plate Equation* (MJBBE or MJBPE) method described in Atmadja and Bagtzoglou (2001), Cornacchiulo and Bagtzoglou (2002), and Bagtzoglou and Atmadja (2003) and then coupled to Discrete Fourier Transform (DFT) processing techniques as originally proposed by Carasso *et al.* (1978) to significantly improve computational efficiency.

MATHEMATICAL APPROACH

The MJBPE method solves first an auxiliary problem by using a new variable w and introducing a stabilization factor s defined as follows:

$$w = e^{st} C \tag{2}$$

$$s = \frac{1}{T} \ln\left(\frac{M}{\delta}\right) \tag{3}$$

where T is the desired reconstruction time duration. Following Carasso's development, δ and M represent the constraints or the acceptable errors on the initial and terminal concentration data in the backward problem that are needed to overcome the issues of stability and non-uniqueness of the solution. They are defined as follows:

$$\left\| C(T_{bi}) - C_2 \right\|_2 \le \delta \tag{4}$$

$$\left\| C(T_{bt}) - C_1 \right\|_2 \le M \tag{5}$$

where T_{bi} and T_{bt} represents the initial and terminal time of the backward problem.

After differentiating equation (2) twice with respect to time and substituting in the original advection-disperion equation (1), we obtain a solvable auxiliary problem defined as:

$$\frac{\partial^2 w}{\partial t^2} = \left(\frac{\partial}{\partial x}\left[D_x \frac{\partial}{\partial x} \right] + \frac{\partial}{\partial y}\left[D_y \frac{\partial}{\partial y} \right] - \frac{\partial}{\partial x}[u] - \frac{\partial}{\partial y}[v] - s \right)^2 w \tag{6}$$

with approriately transformed initial and boundary conditions. Once we solve for $w(x,y,t)$ we apply the following to get the solution sought:

$$C = e^{-st} w \tag{7}$$

The restoration process comprises finding the optimum values of s and K, namely the stabilization factor and the pseudo-coefficient of dispersion that will minimize several possible errors. Since the search for those parameters can be very long, we enhanced the method by automating the search for those parameters. Therefore, we implemented a powerful optimization scheme, commonly referred to as Sequential Quadratic Programming (SQP) to solve our non-linear constrained minimization problem, constrained in a sense that the input variables are bounded. This SQP is also sometimes called an iterative quadratic programming method because it consists of generating a sub-problem that is easier to solve at each iteration. Following is a brief overview of the method; however, the reader is referred to Gill *et al*. (1981) for a more thorough description.

Within each iteration several steps are followed. The first consists of calculating the Hessian matrix of the Lagrangian function using the efficient BFGS Quasi-Newton algorithm (Broyden, 1967; Fletcher, 1970, Goldfarb, 1969; Shanno, 1970). The BFGS procedure uses the evaluation of not only the objective function but also its derivatives calculated by finite difference approximations. This procedure was found more efficient than the Newton algorithm because it is capable of keeping track of information about the steepest descent at each iteration, and hence does not need to re-compute the entire Hessian matrix each time. It is also very closely related to the popular conjugate-gradients methods (Nazareth, 1979). Then the sub-problem is solved using a quadratic programming algorithm for a sequence of feasible points. This step generates an estimate of the search direction solution that is needed to generate the next iteration. A line search procedure is finally involved via the evaluation of a merit function to ensure the convergence of the solution.

In our problem, the input parameters (s and K) are optimized for increasing backward times using an objective function that is set as an equally weighted sum of different mass and point concentration errors. This procedure ultimately generates the time evolution of the optimum parameters s and K. As a result, we are able to reconstruct the 2D profile of the plume at any time step for which values of the pair of optimal parameters are available. Finally, and almost as important, we also obtain the time evolution of the various mass and point errors at the observation or conditioning point(s) as we are recovering the spatial and temporal structure of the plume.

COMPUTATIONAL IMPLEMENTATION

Our algorithm requires only an input array of discrete data, information on how far back one wishes to produce a reconstruction and over how many time steps, some initial guess and bounding constraints for the variables s and K, and finally the number and location of conditioning points. It is worth noting that our program will be able to proceed even if there is no conditioning point available. In this case, mass conservation will be satisfied as the objective function will only be based on the calculation of the mass balance error.

We first uniformly discretize the 2D space domain in both directions and create a $N_g \times N_g$ stiffness matrix. We also discretize the time variable by dividing the backward time interval [0, T] into N_t equally time-spaced interior points. The variable w becomes a discrete variable in space and time and we use the following notation to describe it:

$$w^n = w(x, y, t_n) \qquad \text{for } n=1, 2, \ldots, N_t \qquad (8)$$

If we now defined operator A, such that $A=\nabla^2$ with ∇ being defined as follows:

$$\nabla = \left(\frac{\partial}{\partial x}\left[D_x \frac{\partial}{\partial x} \right] + \frac{\partial}{\partial y}\left[D_y \frac{\partial}{\partial y} \right] - \frac{\partial}{\partial x}[u] - \frac{\partial}{\partial y}[v] - s \right) \qquad (9)$$

we can write the 2D partial differential equation expressed in w using finite difference approximations. The equation becomes then:

$$\frac{\left(w^{n+1} - 2w^n + w^{n-1}\right)}{\Delta t^2} - Aw^n = 0 \qquad (10)$$

with the following transformed boundary conditions:

$$w^0(x, y) = 0 \quad \text{and}$$
$$w^{N_t}(x, y) = e^{st} C(x, y, T) \qquad (11)$$

In matrix form, the transformed equation becomes $\Lambda w=0$ where

$$\Lambda = \begin{bmatrix} -\left(2+A\Delta t^2\right) & I & & & \\ I & -\left(2+A\Delta t^2\right) & I & & \\ & \ddots & \ddots & \ddots & \\ & & I & -\left(2+A\Delta t^2\right) & I \\ & & & I & -\left(2+A\Delta t^2\right) \end{bmatrix} \qquad (12)$$

is a $N_t\ N_g \times N_t\ N_g$ matrix. The overwhelming computational burden is obvious, as even for a reasonable size problem with, for example, a grid size N_g = 20 × 20 = 400 and only 10 marching steps, the matrices involved are formidable (4000 × 4000) and can quickly stress conventional computer abilities. To overcome this problem we incorporate a DFT development as originally proposed by Carasso *et al.* (1978) and first solve the problem in the frequency domain. Once the problem is solved, we then go back into the space domain simply using the inverse Fourier transform.

RESULTS

Test Case 1: One mobile observation or conditioning point

In this section, we analyse the performance of our method using a simplified 2D case. We assume that the coefficient of dispersion is isotropic and spatially invariant. We further assume that the direction amd magnitude of the flow is known *a priori* and is equal in both directions. The observation or conditioning point follows the center of mass of the plume. This case is analogous to a mobile environmental laboratory tracking the plume while transmitting concentration information.

The 2D physical space considered here is a 40 × 40 discretized domain. At t=0, the initial spill is represented by a Dirac function weighted by the mass of the spill M=1, concentrated at one location (ξ, η)=(32, 32). The contaminant then disperses and is advected forward in time following a Gaussian distribution represented by the following analytical solution:

$$C(x,y,t) = \frac{M}{4\pi t D}\exp\left\{ \frac{-(x-\xi+ut)^2}{4Dt} + \frac{-(y-\eta+vt)^2}{4Dt} \right\} \quad (13)$$

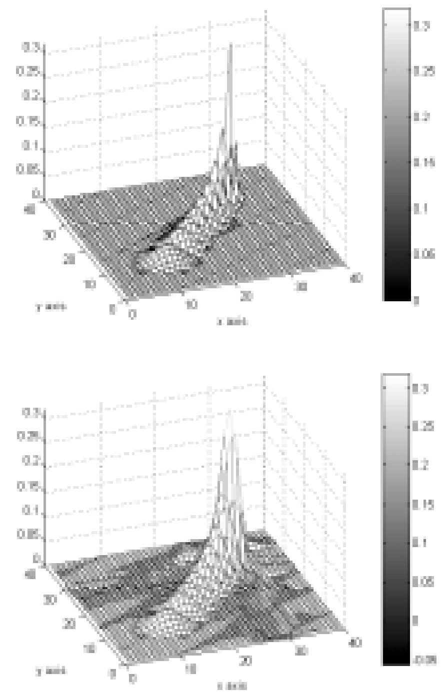
with D_x=D_y=D=0.5 and u=v=4.

A forward simulation is conducted up to t=T_{ft}=5. Figure 1a depicts the evolution of the exact 2D distribution for 8 uniformly spaced time snapshots ranging from 0.5 to 4, and also the distribution corresponding to the forward terminal time at T_{ft}=5. The concentration profile result at $T_{ft}=5$ is then assumed to be the present-day contamination distribution, which is the initial condition for the backward problem.

Applying our algorithm, and using only one conditioning point close to the maximum peak of concentration, we recover the spatial concentration distribution for those 8 different backward times. After normalizing the total time to 1, the recovered concentration corresponds to values of 20% to 90% back in time starting from the initial (t=0) backward time. Figure 1b presents all the recovered distributions so they can be compared to the exact distributions.

These results are further plotted as transect profiles passing through the center of mass for each snapshot and in the x-direction as shown in Figure 2. It is worthwhile noting that the restoration is extremely accurate for up to 60% back in time. After that, even though the peak and mass errors are satisfied, the general shape is corrupted due to spurious undershoots and the corresponding overestimation of the high concentration values.

At each time snapshot the performance of the restoration is also quantitatively evaluated by comparing the forward and backward results and calculating the overall mass error and relative errors at the observation or conditioning point(s). Figure 3 shows that the conservation of mass is satisfied and actually remains within ±0.2% no matter how far back in time we are trying to recover the plume history.



a)

b)

Figure 1: Time evolution of the 2D plume profile. a) Exact distribution and b) Recovered distribution.

Figure 4 shows the recovered concentration versus the exact values at the observation point as a function of backward time. This confirms that the error at this particular conditioning point remains within ±0.05% as required by the optimization program. Therefore, we assert that our algorithm is able to keep track of the increasing concentration of that moving observation or conditioning point as it optimizes for increasing backward times.

We also plot in Figure 5 the evolution of the objective function, or total error, which again clearly shows that mathematically the procedure is working properly by satisfying the imposed constraints.

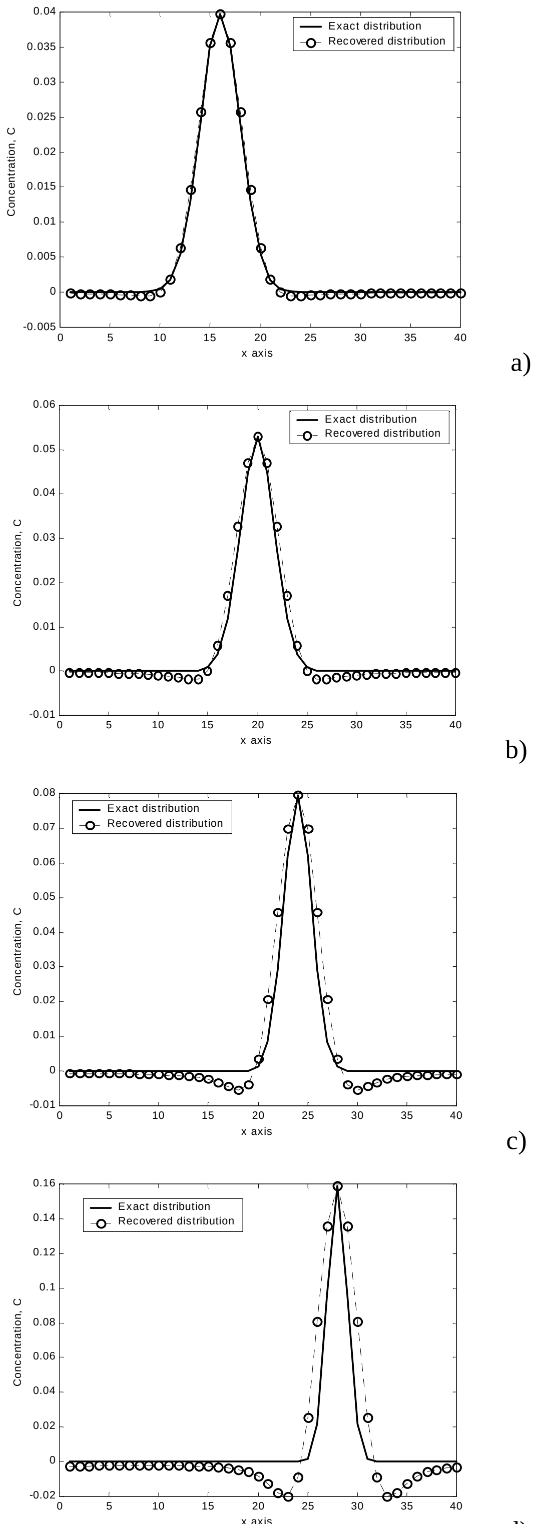


Figure 2: Exact and recovered concentration profiles at a) 20%, b) 40%, c) 60%, and d) 80% backwards in time.

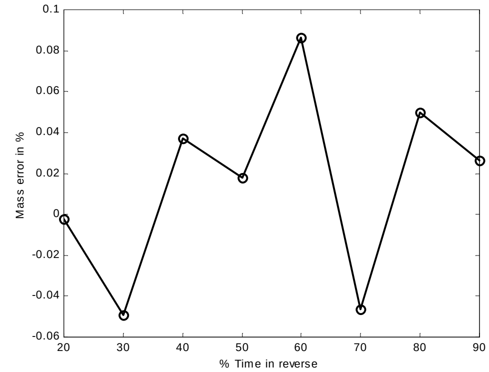


Figure 3: Evolution of the mass error.

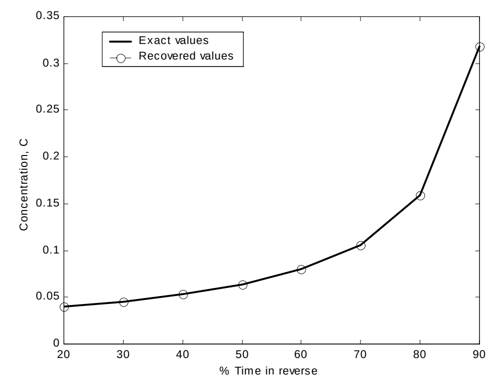


Figure 4: Evolution of the concentration at the single mobile observation or conditioning point.

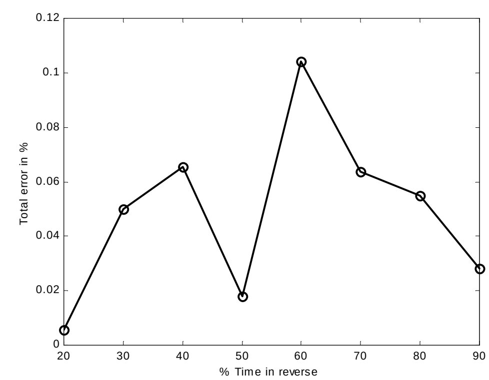


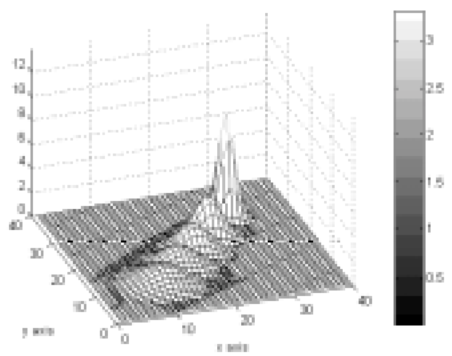
Figure 5: Evolution of the total error, which is a weighted sum of the mass error and the point concentration error.

Test Case 2: Five fixed observation or conditioning points

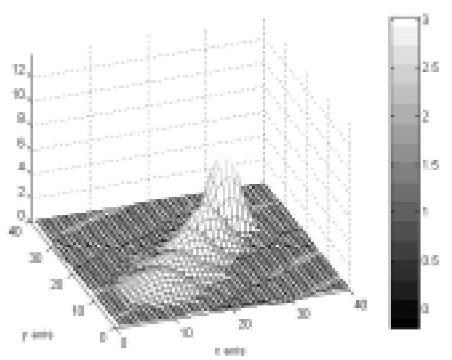
The second case entails a set of observation points that are fixed in space and is analogous to the plume being tracked by a network of bio- or chemical sensors that are stationary. In this case the parameters of the problem are D_x=D_y=D=0.6 and u=v=2.

A forward simulation is conducted up to t=T_{ft}=10. Figure 6a shows the evolution of the exact 2D distribution for 5 snapshots corresponding to forward time of 2, 4, 6, 9, and 10, the last one being to the forward terminal time at T_{ft}=10. The concentration profile result at T_{ft}=10 is then assumed to be the present-day contamination distribution, which is the initial condition for the backward problem.

Applying our algorithm, and using five conditioning points uniformly spaced along the *a priori* known path of the plume, we recover the spatial concentration distribution for various backward times. After normalizing the total time to 1, the recovered concentration corresponds to values of 20% to 90% back in time starting from the initial (t=0) backward time. Figure 6b contains the recovered distribution obtained at the 20%, 40%, 60%, and 85% backward times.



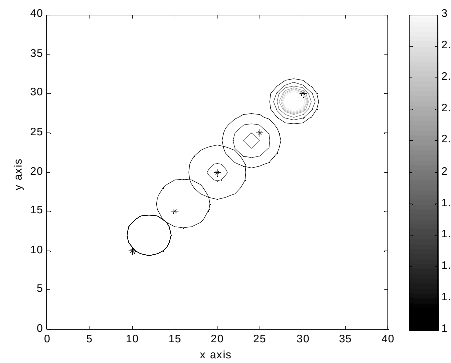
a)



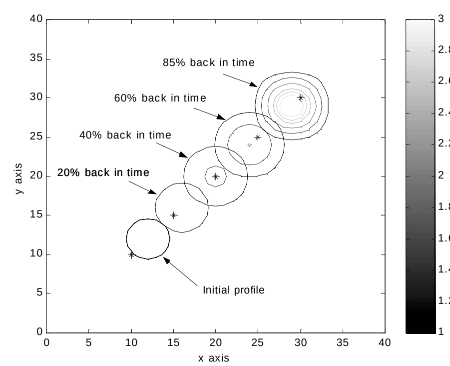
b)

Figure 6: Evolution of the 2D plume profile (initial, 20%, 40%, 60% and 85% back in time). a) Exact distribution and b) Recovered distribution.

A plan view of the plume can be also observed in the form of concentration contours in Figures 7a (exact) and 7b (recovered) with the five observation or conditioning points identified as stars (*). The almost perfect correspondence of the exact and recovered plume's centroid is apparent.



a)



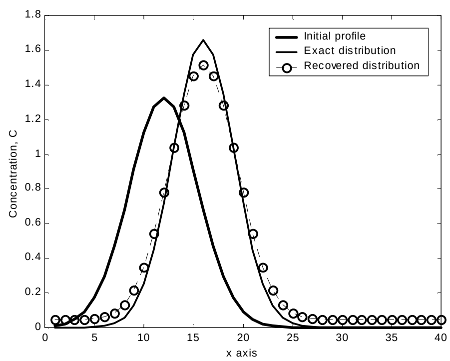
b)

Figure 7: Snapshots of 2D plume in the form of contours (initial, 20%, 40%, 60% and 85% back in time). a) Exact distribution and b) Recovered distribution. Observation points are identified as stars (*) and concentration contours are shown only at the 1, 2, 3, 4, 4.5, 5, 5.5, and 6 levels.

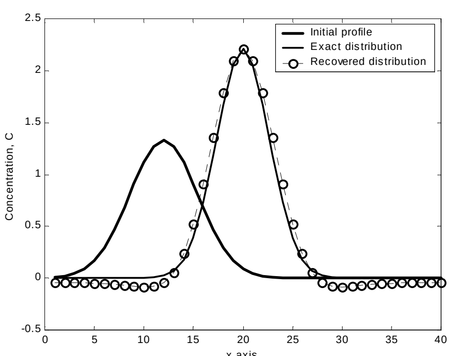
The simulation gave very good results up to a backward time of 65%. After that it became increasingly difficult to balance the error at the conditioning points with the mass balance error as they become more and more "conflicting." The location of the conditioning points relative to the plume is of critical importance, and it would be interesting to analyze how different network configurations can affect the accuracy of the reconstruction results in the future.

To emphasize this point, one can observe in Figure 8 the performance of our reconstruction presented in the form of concentration transect profiles. Of particular interest are Figures 8c, 8d, and 8e.

The reconstruction is considered excellent, very poor, and more than reasonably good for 60%, 80%, and 85% backward times, respectively. The poor performance of the reconstruction for 80% backward time is due to the location of the only two observation or conditioning points that are actively "sensing" the plume at this point in time (points 25, 25 and 30, 30). Because of these points' location with respect to the plume, the method is incapable of improving the reconstruction as both mass balance and point errors are indeed minimal in this case. It is worth noting that this problem is quickly resolved for the 85% backward time as in this case the same observation points make a substantial difference due to their location relative to the plume.



a)



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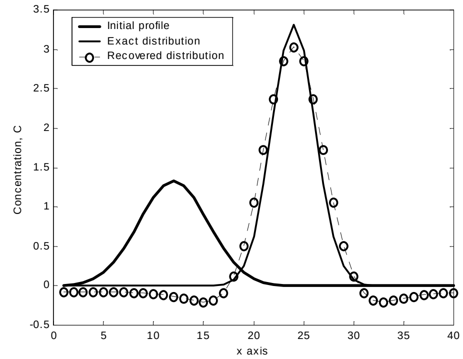
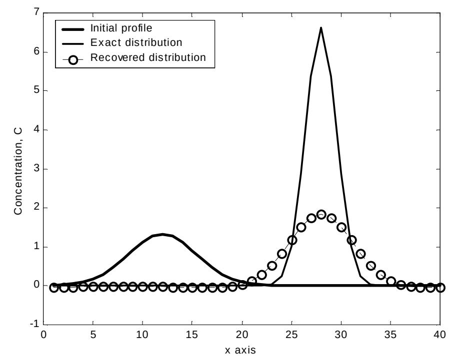
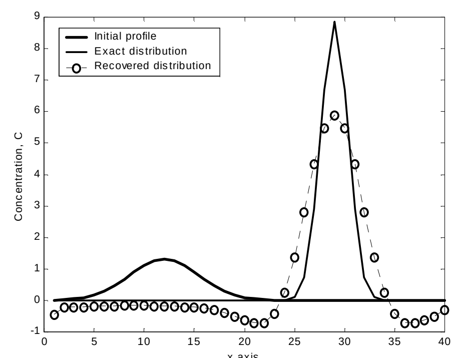


c)



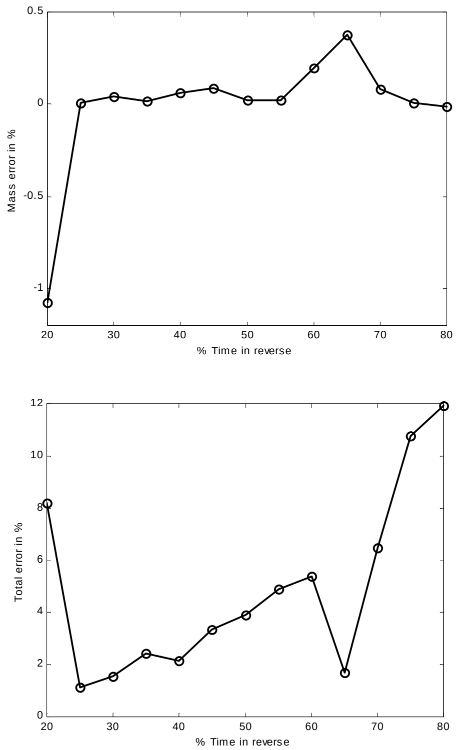
d)



e)

Figure 8: Exact and recovered concentration profiles at a) 20%, b) 40%, c) 60%, d) 80%, and e) 85% backwards in time.

These observations are further supported by Figures 9a and 9b depicting the behavior of the mass balance and total errors as a function of time. Even though the mass balance error is generally less than 0.1% (with an occasional jump at 0.4%), the total error increases dramatically from around 1-5% to 12% for the "problematic" 80% backward time case.



a)

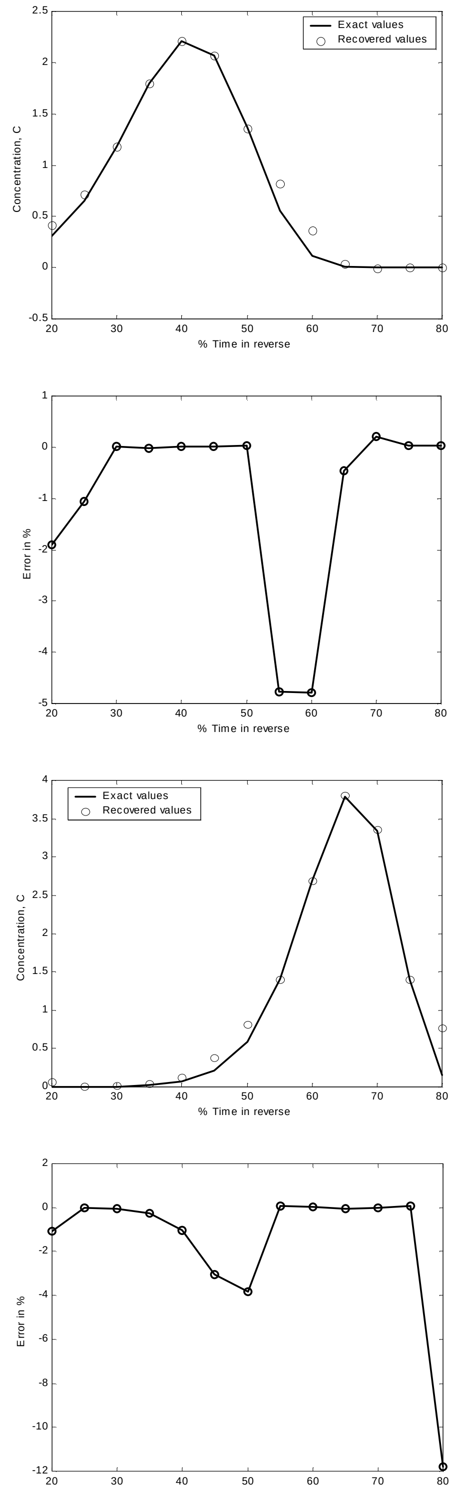
b)

Figure 9: Time evolution of the a) mass balance and b) total error involved in the reconstruction.

Finally, Figure 10 depicts the breakthrough curves, that is concentration versus time curves (10a and 10c) at the two most active of the five observation points (20, 20 and 25, 25) and the associated point concentration error (10b and 10d). Again, the performance of our method is deemed more than satisfactory with the reconstruction producing results that are both accurate (errors averaging less than 2-3%) and retain all salient features of the measured or observed breakthrough curves.

DISCUSSION AND CONCLUSIONS

In this study, we have applied a mathematical method to solve a 2D homogeneous advection-dispersion transport problem backwards in time, and enhanced it by implementing an optimization scheme in order to facilitate and automate the reconstruction process.



a)

b)

c)

d)

Figure 10: Breakthrough curve and corresponding evolution of the error at the conditioning point located at a) and b) (20, 20), and c) and d) (25, 25).

The resulting *Bagtzoglou-Baun* method was then tested and analyzed for two different scenarios. In the first case, our method was capable of reconstructing the time history and location of a non-reactive plume that was instantaneously released. The reconstruction process was based only on a weighted average of the mass error and only one single moving monitoring sensor. That sensor was assumed to be moving along the centroid of the plume and at its known velocity. In the second test case, the objective function was again based on the mass balance error, but this time averaged with errors observed at five fixed monitoring sensors. These sensors were uniformly spaced along the *a priori* known path of the plume.

Several interesting observations can be drawn from our two test cases.

- In the first scenario, the time reconstruction of the 2D plume was successful at every time step, making it clear that even if only one sensor is available, as long as it is close to the centroid of the plume, the contamination source identification algorithm will perform very well.
- In the second scenario, the reconstruction is accurate only until 65% backwards time. In general, the total error increases and the results become inconsistent as we go further backwards in time. We believe that this is due to several factors. First, the position of the sensors relative to where the plume is moving is a critical parameter for a successful reconstruction. Second, the number of sensors is also an important parameter to consider and there may be an ideal number of sensors to be used or activated during the optimization process.

REFERENCES

1. Atmadja, J., and A.C. Bagtzoglou, 2001a, "Pollution source identification in heterogeneous porous media", *Water Resources Research*, 37(8): 2113-2125.
2. Atmadja, J., and A.C. Bagtzoglou, 2001b, "State-of-the-art report on mathematical methods for groundwater pollution source identification", *Environmental Forensics*, 2(3): 205-214.
3. Bagtzoglou, A.C., and J. Atmadja, 2003, "The marching-jury backward beam equation and quasi-reversibility methods for hydrologic inversion: application to contaminant plume spatial distribution recovery", *Water Resources Research*, 39(2): 1038.
4. Broyden, C.G., 1967, "Quasi-Newton methods and their application to function minimization", *Mathematics of Computation*, 21, 368-381.
5. Carasso, A., J.G. Sanderson, and J.M. Hyman, 1978, "Digital removal of random media image degradation by solving the diffusion equation backward in time", *SIAM Journal of Numerical Analysis,* 15(4).
6. Cornacchiulo, D., and A.C. Bagtzoglou, 2002, "The marching-jury backward plate equation for contaminant plume spatial distribution recovery in two-dimensional heterogeneous media: Computational Issues", in *Computational Methods for Subsurface Flow and Transport*, M. Hassanizadeh et al. (eds.), Elsevier Publishers, Netherlands, p. 461-468.
7. Fletcher, R., 1970, "A new approach to variable metric algorithm", *Computer Journal*, 13, 317-322.
8. Gill, E.P, W. Murray, and M. Wright, 1981, *Practical Optimization*, Academic Press, London.
9. Goldfarb, D., 1969, "Extension of Davidon's variable metric method to maximization under linear inequality and equality constraints", *SIAM Journal of Applied Mathematics*, 17, 739-764.
10. Nazareth, L., 1979, "A relationship between the BFGS and conjugate-gradient algorithms and its implications for new algorithm, *SIAM Journal of Numerical Analysis*, 16, 794-800.
11. Shanno, D.F., 1970, "Conditioning of quasi-Newton methods for function minimization", *Mathematics of Computation*, 24, 647-657.