Inverse Modelling for Identifying the Origin and Release rate of Atmospheric Pollution -An Optimisation Approach

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Abstract: This paper describes a mathematical modelling technique which may be used to identify the origin and release history of a polluting gas released into the atmosphere from a point source. The inputs to this model are: pollution concentration measurements made at ground locations downstream, wind speed and transport parameters. The inverse model is formulated as a non-linear least-squares minimisation problem coupled with the solution of an advection-dispersion equation for a non-steady point source. The minimisation problem is ill-posed; consequently its solution is extremely sensitive to errors in the measurement data. Tikhonov's regularisation, which stabilises the solution process, is used to overcome the ill-posedness. Since the minimisation problem has a combination of linear and non-linear parameters, the problem is solved in two steps. Non-linear parameters are found by constructing an iterative procedure and, at each iteration, the linear parameters are calculated. The optimal value of the regularisation parameter is obtained by incorporating the L-curve criterion from linear inverse theory in conjunction with maintaining a steady increase in the regularisation parameter from one iteration to the next. Finally, the accuracy of the model is examined by imposing a normally-distributed relative noise into concentration data generated by the forward model.

Keywords: *Non-linear ill posed problem; Inverse air pollution model; Parameter estimation*

1. INTRODUCTION

Atmospheric dispersion modelling may be used in a post-accident management plan to evaluate conditions in the case where the accidents involve gas leakages. Atmospheric dispersion models describe the transport and dispersal of pollutant gases in the atmosphere. A dispersion model, which is capable of describing the behaviour of air pollutants in the atmosphere, requires the following input data: (1) meteorological data such as wind speed, direction and atmospheric stability, and (2) the source emission rate and its origin. In reality, meteorological data can be measured using available measuring instruments, but the source release rate and its origin are often unknown. Methods to determine the release rate and the origin of the source of the pollutant gas are therefore a significant part of modelling atmospheric dispersion.

The procedure for identifying the origin and release rate of a gas from observations of pollutant concentration reduces to a parameter estimation problem in an air pollution model. Several articles (Edwards *et al.*, 1993; Kibler & Suttles, 1977; Sohier *et al.*, 1997), have been published in this area. In these approaches air pollution transport models for the steady-state point source are used as representative of the pollution transport process, but none of these are based on the advection-dispersion model.

A variety of numerical and analytical techniques have been proposed (Skaggs & Kabala, 1994; Woodbury & Ulrych, 1996; Neupauer *et al*, 2000) to solve similar problems in the area of groundwater modelling. Because of the physical and mathematical similarities between the mass transfer in water and air, mathematical techniques used in groundwater modelling are also relevant to the problem of air pollution modelling.

The novel concept of this study is to identify the origin and release history of a polluting gas based on methods available in the groundwater modelling literature. In one of our previous papers (Kathirgamanathan *et al.*, 2002) we addressed the same problem. In this paper we report a different technique as a computation-

ally more efficient substitute to the previous. The development of this technique is based on the work of Farquharson & Oldenburg (2000).

2. THE FORWARD PROBLEM

A Cartesian co-ordinate system (X, Y, Z) is used with the X-axis oriented in the direction of the mean wind, the Y-axis in the horizontal cross-wind direction, and the Z-axis oriented in the vertical direction. A gas leakage with a mass release rate $q(t)$ kg/s is assumed to start at time $t = 0$ at a point $(0, 0, H)$ height H above the ground, which is assumed horizontal. The released particles are subsequently blown by a wind with mean velocity $\mathbf{u} = (U, 0, 0)$ and monitored at a known location $(X_0, Y_0, 0)$ on the ground. The gas particles move with the wind in the X-direction at the same time as being dispersed by turbulence in the atmosphere. The mass concentration $C(X, Y, Z, t)$ of the cloud of particles is described by (Kathirgamanathan *et al.*, 2002)

$$C(X, Y, 0, t) = \int_0^t K(t, \tau) q(\tau) d\tau, \qquad (1)$$

where the kernel $K(t, \tau)$ is

$$\frac{\exp\left(-\frac{(X-U(t-\tau))^2}{4K_X(t-\tau)} - \frac{Y^2}{4K_Y(t-\tau)} - \frac{H^2}{4K_Z(t-\tau)}\right)}{4\pi^{\frac{3}{2}}(K_X K_Y K_Z)^{\frac{1}{2}}(t-\tau)^{\frac{3}{2}}}, \qquad (2)$$

and K_X, K_Y and K_Z are atmospheric dispersion coefficients in the X, Y and Z directions, respectively.

3. THE INVERSE PROBLEM

Figure 1 illustrates the set-up of the problem. The location and release rate of the pollutant at its source are not available, but the concentration of pollutant distribution $C(X_0, Y_0, 0, t_i) = c_i^P$, $C(X_0 + x_1, Y_0 + y_1, 0, t_i) = c_i^Q$, and $C(X_0 + x_2, Y_0 + y_2, 0, t_i) = c_i^R$ $(i = 1, 2, \ldots, n)$ with $t_1 = 0$, at down-stream locations P, Q, and R respectively are available, where X_0, Y_0 are unknown and x_1, x_2, y_1 and y_2 are known. Here we use the concentration distribution at three points on the ground since it has been demonstrated in one of our previous papers (Kathirgamanathan *et al.*, 2001) that data from at least three spatial locations are necessary to estimate reliably. Our goal here is to estimate the release rate $q(t)$ of the pollution and its location in terms of X_0, Y_0 and H. Here, H is the height of the pollution source from the ground.

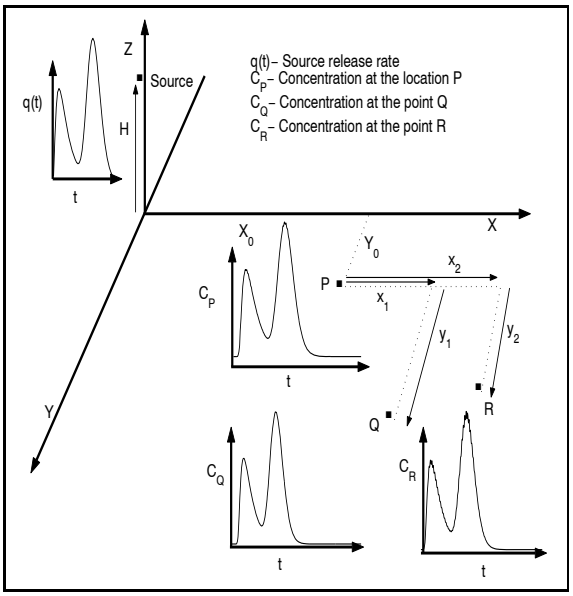

Figure 1. Illustration of the inverse problem.

3.1. The least-squares formulation

The simplest way to proceed is to solve (1) on a time mesh with uniform spacing. We suppose that we wish to determine the source release at times $\tau_1 = 0, \ldots, \tau_m = t_n$, where $m < 3n$ since the number of parameters to be estimated should be no greater than the number n of measurement data points at each location. Discretising (1) and using, for example, the trapezoidal rule gives a system of equations

$$\mathbf{c} = \mathrm{A}(\mathbf{p})\mathbf{q} \qquad (3)$$

where $\mathbf{c} = \left[c_1^P, \ldots, c_n^P, c_1^Q, \ldots, c_n^Q, c_1^R, \ldots, c_n^R\right]^T$, $A_{ij} = [K_P(t_i, \tau_j), K_Q(t_i, \tau_j), K_R(t_i, \tau_j)]^T \beta_{ij}$, $\mathbf{q} = [q(\tau_0), \ldots, q(\tau_m)]^T$ and $\mathbf{p} = [X_0, Y_0, H]^T$, and the β_{ij} are quadrature weights. Generally, minimising an objective function solves inverse problems. Now the problem for estimating the release rate $\mathbf{q}$ and the location $\mathbf{p}$ is

$$\text{minimize} \quad Z(\mathbf{q}, \mathbf{p}) = \|A(\mathbf{p})\mathbf{q} - \mathbf{c}\|_2^2, \qquad (4)$$

where $A(\mathbf{p})\mathbf{q}$, $\mathbf{c}$ are vectors containing the estimated and measured concentrations respectively, $\mathbf{p}$ is the vector of unknown non-linear parameters identifying the source location, and $\mathbf{q}$ is the vector of unknown linear parameters identifying the source release rates. The estimated concentrations are obtained from the solution of the forward problem using estimates

of unknown parameter values.

Since the minimisation problem given in (4) has a combination of linear $\mathbf{q}$ and non-linear parameters $\mathbf{p}$, we separate the solution process into two steps. We find the non-linear parameter $\mathbf{p}$ by constructing an iterative procedure, where at each iteration a linear sub-problem is solved to estimate the linear parameter $\mathbf{q}$ corresponding to that particular value of $\mathbf{p}$. This procedure will speed up enormously because, after the elimination of $\mathbf{q}$ using linear algebra, only three non-linear variables ($\mathbf{p} = [X_0, Y_0, H]$) remain. In one of our previous papers we solved the same problem given in (4) for a known value of $\mathbf{p}$. It was shown that the problem is ill-posed and we therefore used Tikhonov regularisation to solve the problem. The linear sub-problem inside the non-linear iteration is an ill-posed problem.

3.2. Regularized least squares

Tikhonov regularization replaces the ill-posed problem with the well-posed problem by imposing a bound on the solution. With Tikhonov regularisation, we introduce the regularised objective function

$$\begin{aligned} Z(\mathbf{q},\ \mathbf{p}) &= \|A(\mathbf{p})\mathbf{q} - \mathbf{c}\|_2^2 + \lambda^2 \|L\mathbf{q}\|_2^2, \\ &= \phi_d + \lambda^2 \phi_m. \end{aligned} \tag{5}$$

Here, $\phi_d = \|A(\mathbf{p})\mathbf{q} - \mathbf{c}\|_2^2$ is the residual norm (or data misfit function) and $\phi_m = \|L\mathbf{q}\|_2^2$ is the solution norm. We are interested in the function $Z(\mathbf{q}, \mathbf{p})$ and its local and global minima with respect to $(\mathbf{q}, \mathbf{p})$ for different values of the regularisation parameter λ. L is the regularisation operator and λ is the regularisation parameter that controls the relative strength of L, i.e. it compromises between the accuracy and the stability of the solution. The most common form of the regularisation operator is given by

$$\|L\mathbf{q}\|^2 \approx \int_0^{t_n} \left(\frac{d^N q}{d\tau^N} \right)^2 d\tau. \tag{6}$$

The most popular choice for obtaining a smooth solution is $N = 2$ (Skaggs & Kabala 1994).

3.3. Selection of the regularisation parameter

In this section, we build up an algorithm as a computationally more efficient alternative to the algorithm described in our previous paper (Kathirgamanathan *et al.*, 2002). The concept behind this algorithm is based on the work of Farquharson & Oldenburg (2000). In this work, the optimal λ value of the nonlinear minimisation problem

$$\begin{aligned} \min_{\mathbf{x}} \ \phi(\mathbf{x}) &= \|F(\mathbf{x}) - \mathbf{c}\|^2 + \lambda^2 \|L\mathbf{x}\|^2 \\ &= \phi_d(\mathbf{x}) + \lambda^2 \phi_m(\mathbf{x}) \end{aligned} \tag{7}$$

is projected using linear inverse theory (L-curve or GCV). In (7), ϕ_d is an error norm, ϕ_m is a solution norm and λ is a regularisation parameter that balances the two components. In this problem the relationship between the data and the parameters $\mathbf{x}$ is non-linear. The non-linear relationship is overcome by constructing an iterative procedure in which the non-linear minimisation problem is replaced at each iteration by its linearised approximation. In the development of the solution, the value of λ starts from a large value and then decreases from one iteration to the next slowly in conjunction with the equation

$$\lambda_{n+1} = \max(c\lambda_n, \lambda^*), \tag{8}$$

where $0.01 \leq c \leq 0.5$, λ^* is an optimal value of regularisation parameter obtained either using the linear L-curve or the GCV at the n-th iteration for the linearised equation, and λ_n, λ_{n+1} are the values of λ at n-th, $(n+1)$-th iteration, respectively. Equation (8) efficiently imposes a steady decrease on λ values, thereby giving a consistent algorithm for the non-linear inverse problem. Iteration is carried out until λ, the error norm and the solution norm all reach a steady state. At steady state, the optimal value of regularisation parameter, λ^*, obtained using L-curve (or GCV) for the linearised problem, is equal to the final λ value, i.e. $\lambda^* = \lambda_{n+1}$. It has been demonstrated Farquharson & Oldenburg (2000) using simulated data sets that at the final iterations the obtained value of the regularisation parameter was a good estimate of what was expected for a given noise level in the data.

3.4. Some Difficulties

Here, we extend the description of the above approach of estimating the optimal λ to our problem (5). The idea is to solve (5) for a sequences of λ's (each λ acts as the only regularisation parameter for the problem) so that this sequence approaches a steady state as it proceeds. This algorithm is as follows:

1. Choose the starting point $\mathbf{p} = \mathbf{p_0}$, index $I = 1$, and initial value of regularisation parameter $\lambda_I = \lambda_0$, which is very large.

2. Until convergence of λ, $\|A(\mathbf{p})\mathbf{q}-\mathbf{c}\|$, $\|L\mathbf{q}\|$ do

   (i) solve (5) for fixed λ_I, and find the solution $\mathbf{p}=\mathbf{p_I}$;

   (ii) obtain the optimal value of $\lambda=\lambda^*$ for the linear problem

$$\min_{\mathbf{q}} \|A(\mathbf{p_I})\mathbf{q}-\mathbf{c}\|+\lambda^2\|L\mathbf{q}\|$$

   using the linear L-curve;

   (iii) update the λ value, $\lambda_{I+1} = \max(c\lambda_I, \lambda^*)$;

   (iv) take the solution $\mathbf{p_I}$ as the starting point to the next problem with $\lambda = \lambda_{I+1}$;

   (v) I=I+1.

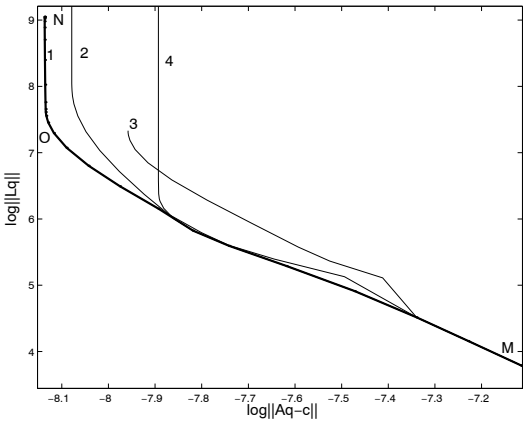

Figure 2. Non-linear L-curves.

By numerical experiments we have found that this approach does not lead to our desired solution. In order to better explain this, we provide the solution path of (5) for a sequence of λ values. Equation (5) contains the error norm (ϕ_d) and the solution norm (ϕ_m) where the first is a non-linear part and the later is quadratic. Equation (5) is non-quadratic if λ is small and therefore the solution of (5) may have several local minima. This leads to several solution paths, as shown in Figure . Suppose that curve 1 is the path of the global minimal solution to (5), and curves 2, 3 and 4 are the corresponding local minimal solutions for various λ values. The value of λ at M is large and therefore gives only one minimum for (5). If we start from M, there is no guarantee the solution path of (5) will be curve 1 in Figure . It might be curves 2 or 3 or 4 in Figure . This example clearly demonstrates that the above approach is not very useful for our problem. To overcome the difficulty, we have to modify this approach slightly.

3.5. The Modified Method

The approach is similar to the method described above, but here we start with a small regularisation parameter and then slowly increase its value. In the first step, we find all or most of the local minima of (5) for the starting value of λ. At this stage we have to be very careful to avoid rank deficiency when we choose a small value of λ.

In the second step, we take each of the local minima obtained from the first step as the starting value to solve (5) for various λ from small to large. We increase λ slowly using the formula $\lambda_{I+1} = \min(c\lambda_I, \lambda^*)$, where $c > 1$, λ^* is the optimal value of the regularisation parameter obtained using the linear L-curve for the sub-problem

$$\min_{\mathbf{q}} \|A(\mathbf{p}^*)\mathbf{q}-\mathbf{c}\|+\lambda^2\|L\mathbf{q}\|,$$

and $\mathbf{p}^*$ is the global minimal solution for λ_I. We repeat this process until λ, the error norm and the solution norm have achieved steady state (or convergence). When c is small, it takes longer to converge on λ, but the gap between two points on the minimal solution curves are close enough and it therefore avoids jumping from one curve to other. Therefore use of a small value for c is recommended.

A sample solution path of minimisation problem (5) is shown in the Figure . In our previous paper (Kathirgamanathan *et al.*, 2002), we solve (5) for a sequence of λ values ($1^{st}\lambda$–$m^{th}\lambda$) and then find the optimal value ($(r+1)^{th}\lambda$). In this paper, we solve (5) for a sequence of λ values from $1^{st}\lambda$ to $(r+1)^{th}\lambda$ only. Therefore, it is clear that some of the work implemented in the previous paper is not necessary. Therefore the method described in this paper is computationally more efficient than the previous method. Both methods are safer since we increase λ slowly, and for each λ we find all local minima and select the minimal solution that gives the objective function minimum. Therefore, there is no way to get trapped in a local minimum. These two methods use different approaches to approximate the optimal value, and therefore each method may find different optimal values of λ.

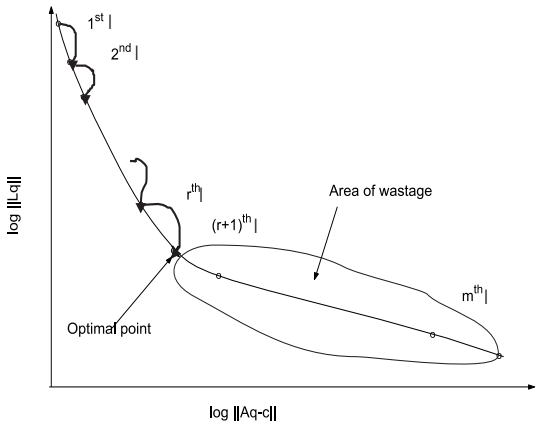

Figure 3. Nonlinear solution path.

4. MODELLING APPLICATION

In this section, we present numerical calculations to evaluate the accuracy of the method developed. To do so, we consider an input of concentration data generated from a point source of strength $q(t)$ $kg\ s^{-1}$ located at $(0, 0, H)$ in the Cartesian coordinate system. We simulate the concentration signals at downstream locations. We obtain concentration signals by using the forward problem (1) and true parameter values. In order to simulate errors, we corrupt the concentration signals by adding normally-distributed random noise. For illustrative purposes, K_X, K_Y, K_Z and U are taken as 12, 12, 0.2113 and 1.8, respectively. The purposes of the numerical example is to demonstrate the simultaneous estimation of parameters X_0, Y_0, H and the source release function $q(t)$. In this example we consider a

Table 1. Estimated values.

p	True values	Estimated values	Confidence interval
X_0	300.0	305.0	$\pm$12.6
Y_0	50.0	47.9	$\pm$9.7
H	12.0	11.4	$\pm$0.7

set of data that is corrupted by 10% of random noise. The results of the source-term estimation are summarised in Table and Figure . Listed in Table are the true non-linear parameter (location) values along with the reconstructed values and their confidence interval estimates. The graph of the regularisation parameter *vs* the number of calculations is shown in Figure a. This figure clearly shows that after a few calculations the regularisation parameter

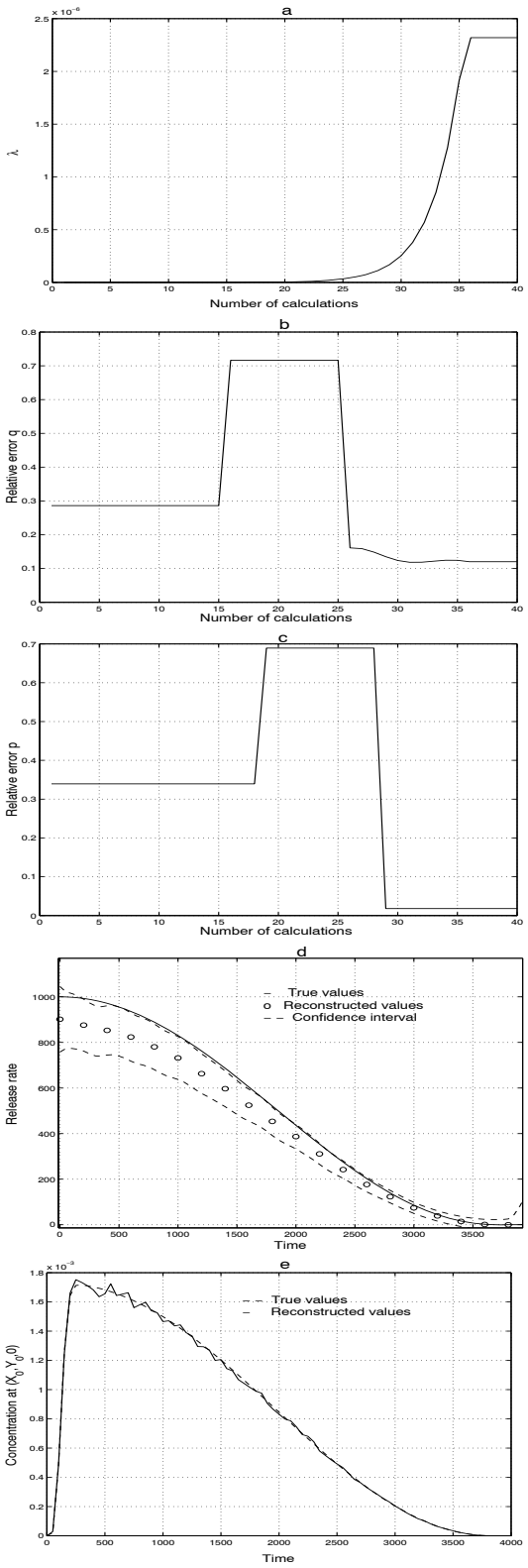

Figure 4. (a) convergence of λ, (b) relative error in $\mathbf{q}$, (c) relative error in $\mathbf{p}$, (d) release rates *vs* time (e) concentration history at the point P=$(X_0, Y_0, 0)$.

converges to 2.34×10^{-6}. Figures b and c depict the error in $\mathbf{q}$ and $\mathbf{p}$ respectively as a function of λ. Figure d depicts the true linear parameters (release rates), along with their reconstructed values and confidence interval estimation. Figure e depicts the true and reconstructed concentration history at the location P. The results from further numerical simulations and comparisons suggest that both approaches perform well, and one particular approach does not always predict a better result than the other approach. In our experience, therefore both approaches are equally useful. If we consider only the amount of computation time, then the approach considered in this paper is a better choice than our previous paper. It is also noted that the accuracy of the estimation decreases with the following: (i) increasing noise in the data, (ii) decreasing the size of the source function discretisation, (iii) regularisation, and (iv) increasing distances between source and observation sites.

5. SUMMARY AND DISCUSSION

The goal of the work presented here is to develop an inverse model capable of simultaneously estimating the location and release rate of a pollutant gas from a point source. The approach is based on a non-linear least squares estimation using pollutant concentration measurements on the ground. As the problem is ill-posed, we apply Tikhonov's regularisation method to stabilise the solution. The parameters in the problem are not all linear. Therefore we determined the linear parameters using simple linear algebra and for the computation of non-linear parameters we then relied exclusively on *MATLAB*'s routine *lsqnonlin*. This process is speeded up enormously because, after the elimination of linear parameters, only a few non-linear parameters remain. The optimal value of the regularisation parameter is obtained using the linear L-curve in conjunction with maintaining a steady increase of the regularisation parameter from one iteration to the next. A numerical example given in the last section shows the inverse model is able to reconstruct the location and release rate of a pollution source with reasonable accuracy.

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