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Application of Bayesian inference methods to inverse modeling for contaminant source identification at Gloucester Landfill, Canada

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A large number of methods are currently available for contaminant source identification, with inverse modeling methods constituting one growing subset. The geostatistical approach to inverse modeling has been shown to be an effective tool for identifying the contaminant release history from a known source in 1-dimensional media [1,2]. In the current work, this approach is extended and used to examine contaminant release history reconstruction for a 3-dimensional plume at the Gloucester landfill site in Ontario, Canada. This work constitutes the first application of methods based on the geostatistical approach to contaminant source identification in a multi-dimensional domain, the first application in a non-point source setting, as well as the first application with both spatially and temporally distributed data. Lastly, a novel approach to enforcing concentration non-negativity is presented and applied.

1. INTRODUCTION

Interest in techniques aimed at identifying sources of environmental contaminants has been growing over the past several years, and a review of such methods is available in [3]. The ability to identify the source of observed contamination can not only help in the remediation process, but can be critical to the identification of responsible parties and to the apportionment of any liability associated with a site. Inverse methods, which constitute one subset, use modeling and statistical tools to estimate the source, taking either a deterministic or stochastic approach [1,4]. The geostatistical approach to inverse modeling is a Bayesian stochastic inference method proposed in the past for the estimation of the release history of a contaminant. Snodgrass and Kitanidis (1997) estimated the release history of a conservative solute in a 1-dimensional homogeneous domain, given point concentration measurements taken after the release [1]. Michalak and Kitanidis (2002) developed a new method for enforcing concentration non-negativity within a geostatistical framework and applied it to the recovery of the groundwater contamination history at Dover Air Force Base, Delaware, using a 1-dimensional diffusion model [2].

In this paper, we infer the 1,4-dioxane release history from the Gloucester landfill in Ontario, Canada, based on downgradient groundwater samples. This work represents the first application of the geostatistical approach to contaminant source identification in a multi-dimensional domain, the first application in a non-point source setting, as well as the first application with both temporally and spatially variable data with . Finally, the non-negativity enforcing method recently developed by [2] is also applied. This dataset has also been examined by [5], using a Minimum Relative Entropy (MRE) approach.

2. SITE DESCRIPTION AND PHYSICAL MODEL

The Gloucester Landfill served as a disposal site for hazardous wastes from 1969 to 1980. These wastes, which were primarily organic solvents, were disposed of in a Special Waste Compound located along the western edge of the landfill. The confined aquifer at the site has been significantly impacted by these wastes. The contaminant exhibiting the greatest mobility at the site is 1,4-dioxane, and the release history was estimated for this compound. The data set consists of measured concentrations in a number of multi-port samplers taken in 1982, and spring/summer 1994, 1995, and 1996. A total of 136 data points are available. The data locations and times are presented in Figure 1, and a subset of the data is presented in Figure 2 along with sampling well locations.

The physical model used for this application was the one used by [5]. The plume is assumed to undergo three-dimensional advective-dispersive transport with steady, uniform groundwater flow. The domain is semi-infinite in the longitudinal direction ($0 \leq x \leq \infty$), infinite in the horizontal-transverse direction ($-\infty \leq y \leq \infty$), and finite in the vertical direction ($0 \leq z \leq B$). Transport is described by the advection-dispersion equation

$$R\frac{\partial c}{\partial t} + V_x\frac{\partial c}{\partial x} - D_x\frac{\partial^2 c}{\partial x^2} - D_y\frac{\partial^2 c}{\partial y^2} - D_z\frac{\partial^2 c}{\partial z^2} = -kc \qquad (1)$$

where c is the aqueous concentration, V_x is the groundwater seepage velocity, D_x, D_y, and D_z are the dispersion coefficients in the longitudinal, transverse horizontal and transverse vertical directions, respectively, R is the retardation factor, and k is a first order biochemical decay constant. The source is modeled as a rectangular Dirichlet patch source at the upstream boundary. Initial and boundary conditions are given by

$$c(x,y,z,0) = 0 \qquad c(0,y,z,t) = \begin{cases} s(t), & -y_0 \leq y \leq y_0, \\ & z_1 \leq z \leq z_2 \\ 0, & \text{otherwise} \end{cases}$$

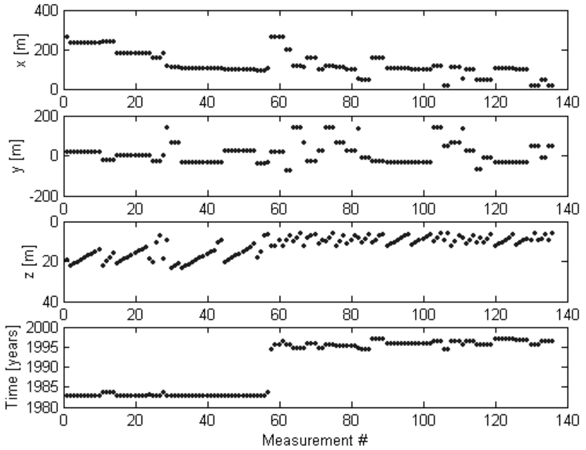

Figure 1. Measurement locations and times.

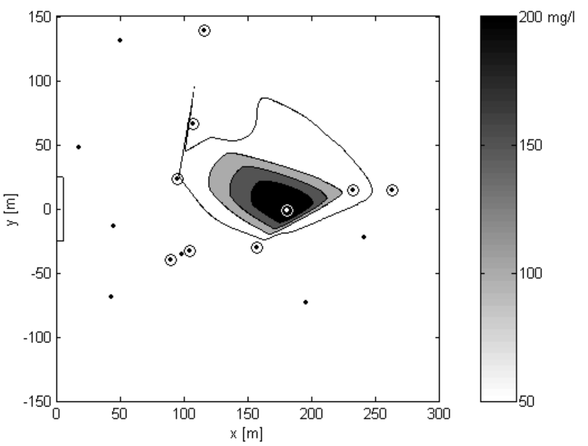

Figure 2. Contour plot of 1,4-dioxane plume based on 1982 measurements (51 data points at 9 locations). The circles represent 1982 measurement locations, the dots represent all measurement locations (1982-1996).

Parameter	V_x	D_x	D_y	D_z	R	k	y_0	z_1	z_2	B
	(m/d)	(m^2/d)	(m^2/d)	(m^2/d)	$(-)$	(d^{-1})	(m)	(m)	(m)	(m)
Value	0.127	0.330	0.064	0.013	1.0	0.0	25.0	6.0	24.0	24.0

Table 1: Physical model parameter values

$$c(\infty, y, z, t) = 0 \qquad c(x, \pm\infty, z, t) = 0 \qquad \left.\frac{\partial c}{\partial z}\right|_{x,y,B,t} = 0 \qquad \left.\frac{\partial c}{\partial z}\right|_{x,y,0,t} = 0 \tag{2}$$

where $s(t)$ is the source concentration, z_1 and z_2 are the upper and lower boundaries of the source, B is the aquifer depth, and y_0 is the half width of the source. The coordinate $(x, y, z) = (0, 0, 0)$ is at the center of the source in the x and y plane, at the top of the aquifer. The required physical parameters were estimated from the 1,4-dioxane plume [5] and are presented in Table 1. The solution to the forward problem is [5]:

$$c(x, y, z, T) = \int_0^T s(t) f(x, y, z, T-t)\, dt \tag{3}$$

$$\begin{aligned} f(x, y, z, T-t) = {} & \tfrac{x(z_2-z_1)}{4B(\pi D_x)^{1/2}} \tfrac{1}{(T-t)^{1.5}} \exp\left[-\tfrac{(x-V_x(T-t))^2}{4D_x(T-t)} - \tau(T-t)\right] \\ & \cdot \left[\operatorname{erfc}\left(\tfrac{y-y_0}{2(D_y(T-t))^{1/2}}\right) - \operatorname{erfc}\left(\tfrac{y+y_0}{2(D_y(T-t))^{1/2}}\right)\right] \\ & + \tfrac{x}{(4D_x)^{1/2}\pi^{1.5}} \tfrac{1}{(T-t)^{1.5}} \exp\left[-\tfrac{(x-V_x(T-t))^2}{4D_x(T-t)} - \tau(T-t)\right] \\ & \cdot \left[\operatorname{erfc}\left(\tfrac{y-y_0}{2(D_y(T-t))^{1/2}}\right) - \operatorname{erfc}\left(\tfrac{y+y_0}{2(D_y(T-t))^{1/2}}\right)\right] \\ & \cdot \sum_{l=1}^{\infty} \tfrac{1}{l} \left[\sin\left(\tfrac{l\pi z_2}{B}\right) - \sin\left(\tfrac{l\pi z_1}{B}\right)\right] \cos\left(\tfrac{l\pi z}{B}\right) \exp\left[-\tfrac{l^2\pi^2 D_z(T-t)}{B^2}\right] \end{aligned} \tag{4}$$

3. INVERSE MODEL

When the governing equation is linear in the unknown, the estimation problem can be expressed as $\mathbf{z} = \mathbf{Hs} + \boldsymbol{\varepsilon}$, where $\mathbf{z}$ is an $n \times 1$ vector of observations, $\mathbf{s}$ is an $m \times 1$ vector of the discretized unknown function, $\mathbf{H}$ is a known sensitivity matrix, and $\boldsymbol{\varepsilon}$ is the measurement error. Following geostatistical methodology, $\mathbf{s}$ and $\boldsymbol{\varepsilon}$ are represented as random vectors. Let $\mathbf{x}_i = [x_i, y_i, z_i], i = 1, \ldots, n$ be the n points at which measurements are taken at times T_i. Let us discretize the time domain into m points $t_j, j = 1, \ldots, m$, with a time step $\Delta t = (t_m - t_1)/(m-1)$. In this case, the sensitivity matrix is:

$$\mathbf{H} = \Delta t \begin{bmatrix} f(\mathbf{x}_1, T_1 - t_1) & \cdots & f(\mathbf{x}_1, T_1 - t_m) \\ f(\mathbf{x}_2, T_2 - t_1) & \cdots & f(\mathbf{x}_2, T_2 - t_m) \\ \vdots & \ddots & \vdots \\ f(\mathbf{x}_n, T_n - t_1) & \cdots & f(\mathbf{x}_n, T_n - t_m) \end{bmatrix} \tag{5}$$

We assume that $\boldsymbol{\varepsilon}$ has zero mean and known covariance matrix $\mathbf{R} = \sigma_R^2 \mathbf{I}$, where σ_R^2 is the variance of the measurement error, and $\mathbf{I}$ is an $n \times n$ identity matrix. For this problem, a linear but unknown trend in the contaminant release concentration was assumed. Thus,

$$E[\mathbf{s}] = \mathbf{Y}\boldsymbol{\beta} \qquad \mathbf{Y} = \begin{bmatrix} 1 & \cdots & 1 \\ t_1 & \cdots & t_m \end{bmatrix}' \tag{6}$$

where $\mathbf{Y}$ is a known $m \times p$ matrix and $\boldsymbol{\beta}$ are p unknown drift coefficients. The covariance function of $\mathbf{s}$ was assumed to have cubic form:

$$Q\left(t_i, t_j|\theta\right) = \theta \left|t_i - t_j\right|^3 \tag{7}$$

where $|t_i - t_j|$ is the separation distance (in units of time) and θ is an unknown parameter.

The structural parameters, in this case θ and σ_R^2, are estimated by maximizing the probability of the measurements [6]:

$$p\left(\mathbf{z}|\theta, \sigma_R^2\right) \propto |\boldsymbol{\Sigma}|^{-1/2} \left|\mathbf{Y}^T\mathbf{H}^T\boldsymbol{\Sigma}^{-1}\mathbf{H}\mathbf{Y}\right|^{-1/2} \exp\left[-\frac{1}{2}\mathbf{z}^T\boldsymbol{\Xi}\mathbf{z}\right] \tag{8}$$

$$\boldsymbol{\Sigma}{=}\mathbf{H}\mathbf{Q}\mathbf{H}^T + \mathbf{R} \qquad \boldsymbol{\Xi}{=}\boldsymbol{\Sigma}^{-1} - \boldsymbol{\Sigma}^{-1}\mathbf{H}\mathbf{Y}\left(\mathbf{Y}^T\mathbf{H}^T\boldsymbol{\Sigma}^{-1}\mathbf{H}\mathbf{Y}\right)^{-1}\mathbf{Y}^T\mathbf{H}^T\boldsymbol{\Sigma}^{-1} \tag{9}$$

where $|\ |$ denotes matrix determinant.

Once these parameters have been estimated, the optimization problem becomes:

$$p''\left(\mathbf{s}\right) \propto \exp\left[-\frac{1}{2}\left(\left(\mathbf{z} - \mathbf{H}\mathbf{s}\right)^T\mathbf{R}^{-1}\left(\mathbf{z} - \mathbf{H}\mathbf{s}\right) + \left(\mathbf{s} - \mathbf{Y}\boldsymbol{\beta}\right)^T\mathbf{Q}^{-1}\left(\mathbf{s} - \mathbf{Y}\boldsymbol{\beta}\right)\right)\right] \tag{10}$$

$$\mathbf{z} = \left[\begin{array}{cccc} z\left(\mathbf{x}_1, T_1\right) & z\left(\mathbf{x}_2, T_2\right) & \cdots & z\left(\mathbf{x}_n, T_n\right)\end{array}\right]' \qquad \mathbf{s} = \left[\begin{array}{cccc} s\left(t_1\right) & s\left(t_2\right) & \cdots & s\left(t_m\right)\end{array}\right]' \tag{11}$$

The corresponding system to be solved is:

$$\left[\begin{array}{ccc} \boldsymbol{\Sigma} & \vdots & \mathbf{H}\mathbf{Y} \\ \cdots & & \cdots \\ \left(\mathbf{H}\mathbf{Y}\right)^T & \vdots & 0 \end{array}\right] \left[\begin{array}{c} \boldsymbol{\Lambda}^T \\ \cdots \\ \mathbf{M} \end{array}\right] = \left[\begin{array}{c} \mathbf{H}\mathbf{Q} \\ \cdots \\ \mathbf{Y}^T \end{array}\right] \tag{12}$$

where $\boldsymbol{\Lambda}$ is a $m \times n$ matrix of coefficients and $\mathbf{M}$ is a $p \times m$ matrix of multipliers. The best estimate of the function and its covariance are

$$\widehat{\mathbf{s}} = \boldsymbol{\Lambda}\mathbf{z} \qquad \mathbf{V}_{\widehat{\mathbf{s}}}{=}{-}\mathbf{Y}\mathbf{M} + \mathbf{Q} - \mathbf{Q}\mathbf{H}^T\boldsymbol{\Lambda}^T \tag{13}$$

Using geostatistical methodology, it is also possible to generate realizations of the release history that are conditional on all the observations [6,7]. First, an unconditional unconstrained realization $\mathbf{s}_{uu,l}$ and a realization of the error vector $\boldsymbol{\varepsilon}_l$ are generated. Then, the conditional unconstrained realization $\mathbf{s}_{cu,l}$ is found by minimizing

$$J = \left(\mathbf{s}_{cu,l} - \mathbf{s}_{uu,l}\right)^T\mathbf{G}\left(\mathbf{s}_{cu,l} - \mathbf{s}_{uu,l}\right) + \left(\mathbf{z} + \boldsymbol{\varepsilon}_l - \mathbf{H}\mathbf{s}_{cu,l}\right)^T\mathbf{R}^{-1}\left(\mathbf{z} + \boldsymbol{\varepsilon}_l - \mathbf{H}\mathbf{s}_{cu,l}\right) \tag{14}$$

$$\mathbf{G} = \mathbf{Q}^{-1} - \mathbf{Q}^{-1}\mathbf{Y}\left(\mathbf{Y}^T\mathbf{Q}^{-1}\mathbf{Y}\right)^{-1}\mathbf{Y}^T\mathbf{Q}^{-1} \tag{15}$$

with respect to $\mathbf{s}_{cu,l}$, where the subscript l denotes the lth realization.

We also apply a novel non-negativity enforcing method [2] to the data. A Metropolis-Hastings algorithm is used with conditional realizations generated using a cubic semi-variogram, modified to be nonnegative using Lagrange multipliers. The unconditional realizations used to obtain the candidate conditional realizations are sequentially correlated:

$$\mathbf{s}_{uu,c} = \phi\mathbf{s}_{uu,l} + \alpha\mathbf{u}_c \qquad 0 < \phi < 1, \qquad \alpha = \sqrt{1 - \phi^2} \tag{16}$$

where $\mathbf{u}_c$ is an independently generated unconditional realization, and $\mathbf{s}_{uu,l}$ is the unconditional realization used in the generation of the last accepted realization. A conditional unconstrained realization $\mathbf{s}_{cu,c}$ is obtained from $\mathbf{s}_{uu,c}$ using the geostatistical procedure described earlier, and the candidate conditional constrained realization $\mathbf{s}_{cc,c}$ is obtained by applying the method of Lagrange multipliers to $\mathbf{s}_{cu,c}$. The probability of acceptance of $\mathbf{s}_{cc,c}$ is:

$$\varsigma\left(\mathbf{s}_{cc,c}|\mathbf{s}_{cc,l}\right) = \min\left\{\frac{p''\left(\mathbf{s}_{cc,c}\right) q\left(\mathbf{s}_{cc,l}|\mathbf{s}_{cc,c}\right)}{p''\left(\mathbf{s}_{cc,l}\right) q\left(\mathbf{s}_{cc,c}|\mathbf{s}_{cc,l}\right)}, 1\right\} \tag{17}$$

where $\mathbf{s}_{cc,l}$ is the last accepted conditional constrained realization, $p''\left(\mathbf{s}_{cc,\cdot}\right)$ is the posterior probability distribution as defined in Eqn. 10, and $q\left(\mathbf{s}_{cc,\cdot}|\mathbf{s}_{cc,\cdot}\right)$ is the transition probability from one realization to the other. We approximate $q\left(\mathbf{s}_{cc,\cdot}|\mathbf{s}_{cc,\cdot}\right)$ by $q\left(\mathbf{s}_{uu,\cdot}|\mathbf{s}_{uu,\cdot}\right)$, the transition probability from one unconditional realization to the other. For example,

$$q\left(\mathbf{s}_{uu,c}|\mathbf{s}_{uu,l}\right) \propto \exp\left[-\frac{1}{2}\left(\mathbf{s}_{uu,c} - \phi\mathbf{s}_{uu,l}\right)^T \frac{\mathbf{Q}^{-1}}{\alpha^2}\left(\mathbf{s}_{uu,c} - \phi\mathbf{s}_{uu,l}\right)\right] \tag{18}$$

A number u is sampled from a uniform distribution in the range $[0, 1]$. If this number is less than $\varsigma\left(\mathbf{s}_{cc,c}|\mathbf{s}_{cc,l}\right)$ then $\mathbf{s}_{cc,l+1} = \mathbf{s}_{cc,c}$, otherwise, $\mathbf{s}_{cc,l+1} = \mathbf{s}_{cc,l}$. The chain is run until the probability space has been appropriately sampled.

4. RESULTS AND DISCUSSION

The data were sorted in ascending order of maximum sensitivity time, defined as:

$$t_{i,o} = T_i - x_i/V_x \tag{19}$$

This is the release time for which the sensitivity of the measurement is maximized. This has no effect on the source estimation, but is useful for presenting data and results.

The results presented in Figure 3 were obtained by applying the linear unconstrained geostatistical methodology. The measurement error variance σ_R^2 was estimated to be $1.28\times10^{-2}\ (mg/l)^2$ and the parameter θ used in the cubic variogram model was estimated to be $1.44\times10^{-9}\ mg^2/\left(l^2 day^3\right)$. The standard deviation of data reproduction is $0.110\ mg/l$ ($\sigma^2 = 1.20 \times 10^{-2}\ (mg/l)^2$), which is consistent with the estimated σ_R^2.

Figure 3 suggests that the precision with which the release history can be estimated varies significantly with time. Whereas the confidence intervals about the best estimate for the periods 1977-1981 and 1988-1996 are fairly narrow, they are much wider for other periods. Figure 4 presents the sensitivity matrix $\mathbf{H}$ of the measurement data to releases that would have occurred at given times. This figure confirms that the data only supply information about the release history over a given period. Measurements 1-57 jointly define the release history from 1977 through 1981, and measurements 58-136 provide information for 1988 through 1996. These observations point out one of the advantages of using a stochastic approach such as MRE or Bayesian inference: it provides not only a best estimate, but also confidence intervals which are representative of the information contained in the available data.

Woodbury et al. (1998) stated that the "best interpretation obtainable from this modelling exercise suggests that (1) it is not possible to resolve the source release history prior

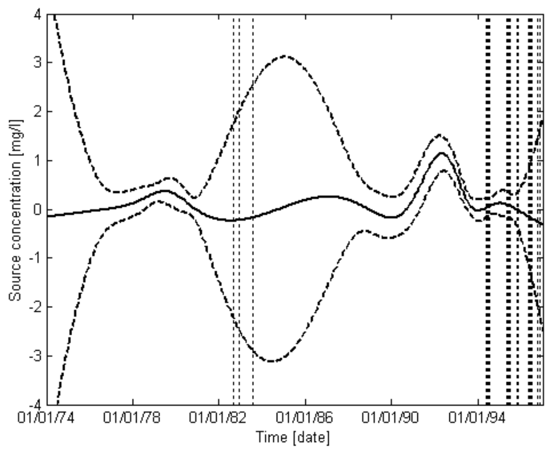

Figure 3. Recovered release history using linear geostatistical inversing. Solid line represents the best estimate, dashed lines the upper and lower 95% confidence intervals, and vertical dotted lines times at which measurements were collected.

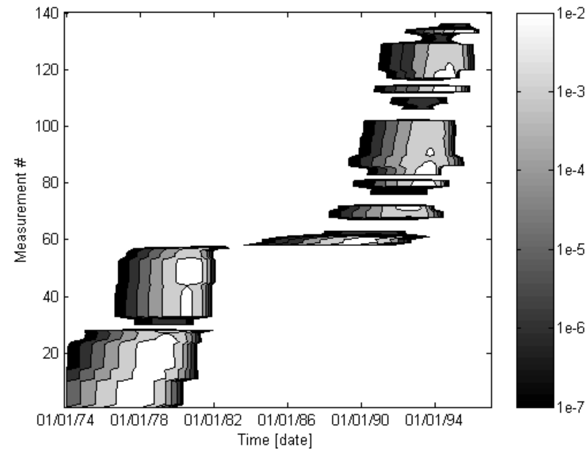

Figure 4. Sensitivity matrix **H** of measurements to release times (note logarithmic scale).

to 3200 days [February 1978] and after about 4400 days [May 1981], and (2) there is a 90% probability that the 'true' source release lies within the range of about 0.1 to 2 mg/l between those dates" [5]. Our results confirm that the precision with which the source can be estimated drops off significantly prior to 1977 and after 1981. During this well-defined period, the 95% confidence intervals are less than $0.6\,mg/l$ wide. A determination can also be made, however, regarding the release history in the period 1988-1996, with confidence intervals being less than $1.0\,mg/l$ in width. These last results indicate that contaminant release continued after the end of landfill operations. Furthermore, Woodbury et al. (1998) aimed only to estimate the release history from 1976 through 1980 [5], and used all available data, including those collected in 1994-1996 (measurements 58 - 136). These later data, however, give no information about the release history prior to 1987.

The results presented in Figure 5 were generated by applying the non-negativity enforcing methodology developed by [2], using 50,000 realizations. The values of σ_R^2 and θ used were those from the solution of the unconstrained problem. Figure 6 shows three sample conditional realizations, illustrating the variety of possible release scenarios. The standard deviation of data reproduction of the best estimate is $0.111\,mg/l$ ($\sigma^2 = 1.23 \times 10^{-2}\ (mg/l)^2$). These results are generally consistent with those presented in Figure 3, but have the advantage of representing a physically significant release history best estimate and confidence intervals, without sacrificing data reproduction. The peak concentration values for the two periods for which the release history is well defined are not significantly affected by the addition of the non-negativity constraint. The 95% confidence intervals are somewhat narrower, being less than $0.5\,mg/l$ wide for the period 1977-1981 and $0.8\,mg/l$ wide for 1988-1996.

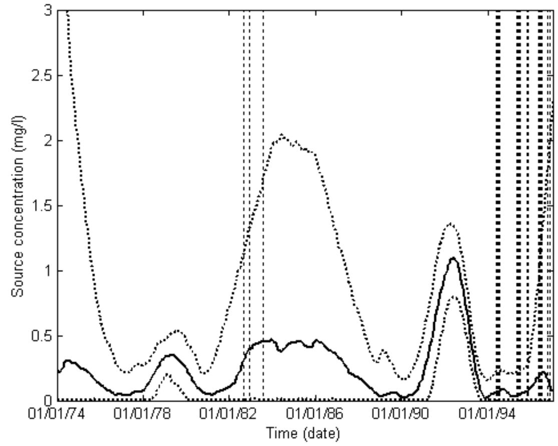

Figure 5. Recovered release history using non-negativity enforcing method. Solid line represents the best estimate, dashed lines the upper and lower 95% confidence intervals, and vertical dotted lines times at which measurements were collected.

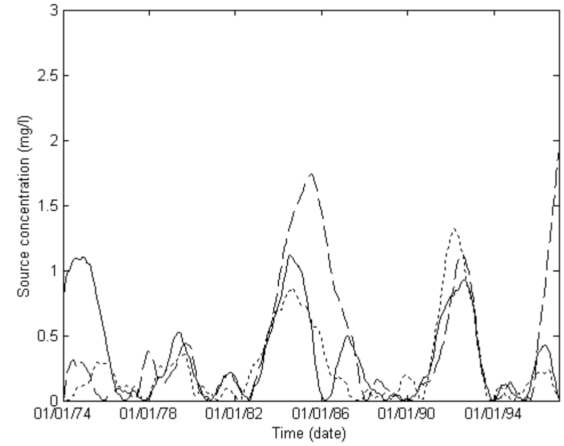

Figure 6. Conditional realizations of release history using non-negativity enforcing method.

For the intermediate period, however, the non-negativity enforcing method shows a significant positive estimated source, whereas the linear geostatistical method does not. This discrepancy is due to the wide confidence intervals, and corresponding low precision with which the release history can be defined for this time period. If nothing is known about the release history for a given period, the linear method will yield a flat estimate close to the levels observed at the boundaries of well defined regions. In this case, this corresponds to estimates around zero. For the non-negativity enforcing method, the estimate for this period is offset away from zero, because the only available information is that the source release history for that period is non-negative. This discrepancy between the results obtained using the linear and non-negativity enforcing methods should not be misleading, however, if one observes the width of the confidence intervals. Both methods yield wide confidence intervals for this period, indicating that the best estimate should not be taken as a clear indication of the actual release history for this period, but instead as the median of the wide range of possible release histories, given the lack of information about the release during that period.

The MRE and geostatistical approaches introduce prior information in different ways, which accounts for some differences between current results and those presented in [5]. In both methods, information contained in the prior is combined with information supplied by the data to yield a pdf for the release function. The prior information for the MRE method is supplied by specifying bounds on the function and using a truncated exponential pdf to form the prior pdf. In addition, a prior mean is specified at each point. For example, in [5], a Gaussian prior centered in 1978 with a range of 600 days as well as a uniform prior from early 1978 to late 1980 were used, both with given concentration levels. Therefore, an estimate of the form and value of the unknown function is supplied as a prior. The assumptions made in the geostatistical approach are less restrictive. Basically, the way in which the unknown function varies in space or time is assumed. For example, using a linear variogram corresponds to the assumption that the function varies erratically at small scales, but that the smallest estimate is sought, whereas using a cubic variogram yields flat and smooth estimates, minimizing the second derivative of the estimates. In the current work, the prior mean was assumed to be a linear trend, with a-priori unspecified parameters, and even the prior variance used with these variograms is estimated using the data themselves. Therefore, the geostatistical approach uses less stringent assumptions in the manner in which it introduces prior information into the system.

Contrary to current results, Woodbury et al. (1998) obtained distributions with peaks at the earliest and latest times within the range of their prior distributions. However, these peaks may be a product of the prior being used. If a release occurring over a wider time range than that supported by the prior is required to reproduce observations, then the estimated release history at the earliest and latest times supported by the prior will be high, as the model attempts to reproduce measurements that are actually most sensitive to releases that occurred at times for which the prior forces the release to be zero.

5. CONCLUSIONS

Using 136 measurements taken downgradient from the Special Waste Compound at Gloucester Landfill in Ontario, Canada, we estimated the release history of 1,4-dioxane from this facility. A linear geostatistical method was applied, as was a novel concentration non-negativity enforcing approach. The available data allow for relatively precise release estimation for 1977-1981 and 1988-1996, with 95% confidence intervals less than $1.0\,mg/l$ wide. The estimates reproduce the observations to within the estimated measurement error variance, which takes into account both analytical measurement error and the limitations of the physical model. Results are generally consistent with those reported in [5], where a Minimum Relative Entropy approach was applied to the dataset.

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