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Backward probabilistic model of groundwater contamination in non-uniform and transient flow

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Abstract

Backward location and travel time probabilities, which provide information about the former location of contamination in an aquifer, can be used to identify unknown contamination sources. Backward location probability describes the possible upgradient positions of contamination at a known time in the past, and backward travel time probability describes the time required for contamination to travel from a known upgradient location to an observation point. These probabilities are related to adjoint states of resident concentration, and their governing equation is the adjoint of a forward contaminant transport model. Using adjoint theory to obtain the appropriate governing equation, we extend the backward probability model for conservative solutes to more general non-uniform and transient flow fields. In particular, we address three important extensions, spatially-varying porosity, transient flow and temporally-varying porosity, and internal distributed sources and sinks of solute and water. For the first time we learn that forward and backward location and travel time probabilities are not necessarily equivalent to adjoint states, but are related to them. The extensions are illustrated using a vertically-integrated groundwater model, creating transient flow by a step change in pumping and using areal recharge as an internal distributed source. Both the movement and spread of probabilities are affected. With internal sources of water, there are two interpretations of backward probability, depending on whether or not the source of water is also a source of solute. The results demonstrate how the backward probability model can be applied to other, perhaps more important, non-uniform and transient flow conditions, with time- and space-varying water storage, such as time-varying pumping or unsaturated (or saturated–unsaturated) flow and transport with spatially- and temporally-varying moisture content.

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1. Introduction

When contamination is observed in an aquifer, the source of contamination is often unknown. To remediate the aquifer or to assign responsibility we might need to identify the contamination source or estimate the release time of contamination from a known source. Since the source is unknown, this information must be inferred from the observed distribution of the contamination using knowledge of the contaminant transport processes. For example, one could select several possible sources of contamination and individually simulate the evolution of a contaminant plume from each of these sources, comparing the simulation results with the measured data. This approach is computationally inefficient because one simulation must be run for each source, and the results are limited to only the preselected sources.

Backward modeling [16,17,23,24] is a more efficient approach that can be used to address these and related issues, such as well head protection and aquifer vulnerability. This method produces backward location or travel time probability distributions that can be used to characterize the source location and source release time. Backward location probability is a probability distribution describing the possible prior positions of the contamination (or possible source locations); and backward travel time probability describes the time for a solute parcel to travel from an upgradient location

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(possibly the source location) to the observation point. These probabilities are related to marginal sensitivities. This can be seen most clearly by first examining their forward counterparts, forward location and travel time probabilities, which are often used in forward modeling to describe solute transport in groundwater [2–4,10,12].

Forward location probability, $f_{\mathbf{x}}(\mathbf{x}; t, \mathbf{x}_0)$, describes the random position, $\mathbf{x}$, of a solute parcel at a fixed time, t, after its release from a known source at $\mathbf{x}_0$ [2–5,12] and is related to resident concentration [4,12]. For contamination originating from an instantaneous point source at $\mathbf{x}_0$, the probability of finding a solute parcel in a control volume centered at $\mathbf{x}$ at time T is equivalent to the ratio of the solute mass in that volume to the total mass of solute in the aquifer; therefore, the location probability density function at forward time $t = T$, is given by

$$f_{\mathbf{x}}(\mathbf{x}; t = T, \mathbf{x}_0) = \frac{\theta(\mathbf{x}, T) C(\mathbf{x}, T)}{M_0}, \tag{1}$$

where $f_{\mathbf{x}}(\mathbf{x}; t = T, \mathbf{x}_0)$ is location probability at time T, $\mathbf{x}$ is the position vector (random variable), $C(\mathbf{x}, T)$ is the distribution of resident concentration at all $\mathbf{x}$ at time T due to the instantaneous point source, $\mathbf{x}_0$ is the known source location, M_0 is the source mass, and θ is porosity. Porosity may vary in space due to aquifer heterogeneity and in space/time due to matrix compressibility, precipitation/dissolution, or other processes. (In unsaturated flow, θ would represent space/time variability of moisture content.) Rearranging (1), we see that $C(\mathbf{x}, T) = (M_0/\theta) f_{\mathbf{x}}(\mathbf{x}; t = T, \mathbf{x}_0)$, leading to $\mathrm{d}C(\mathbf{x}, T)/\mathrm{d}M_0 = C(\mathbf{x}, T)/M_0$, so the forward location probability can also be given by

$$\begin{aligned} f_{\mathbf{x}}(\mathbf{x}; t = T, \mathbf{x}_0) &= \theta(\mathbf{x}, T) \frac{\mathrm{d}C(\mathbf{x}, T)}{\mathrm{d}M_0} \\ &= \theta(\mathbf{x}, T) \psi_x(\mathbf{x}, t = T; \mathbf{x}_0), \end{aligned} \tag{2}$$

where $\psi_x(\mathbf{x}, t = T; \mathbf{x}_0)$ is the state sensitivity of resident concentration at $\mathbf{x}$ to the source mass, M_0, at $\mathbf{x}_0$. Forward location probability at $\mathbf{x}$ is proportional to this sensitivity, and to the porosity at the selected position, $\mathbf{x}$, and at time $t = T$. In the special case of constant porosity, the sensitivity can be taken with respect to $M' = M_0/\theta$, the measure of source mass used in [17]. Note that if there are multiple sources $\mathbf{x}_0$, each has its own forward location probability.

Through a similar approach we see that forward travel time probability is related to a different sensitivity. Forward travel time probability, $f_{\mathrm{t}}(t; \mathbf{x}, \mathbf{x}_0)$, describes the time required (random variable) for a solute parcel to travel from its source at $\mathbf{x}_0$ to a fixed location of interest, $\mathbf{x}$ [5,6,10,11], and is related to flux concentration [19,21]. For an instantaneous point source of contamination at $\mathbf{x}_0$, the travel time probability density function at a specific downgradient location, $\mathbf{x} = \mathbf{x}_{\mathrm{w}}$, is equivalent to the ratio of mass flow rate at $\mathbf{x}_{\mathrm{w}}$ to the total mass in the aquifer, given by

$$f_{\mathrm{t}}(t; \mathbf{x} = \mathbf{x}_{\mathrm{w}}, \mathbf{x}_0) = \frac{|q(\mathbf{x}_{\mathrm{w}}, t)| A(\mathbf{x}_{\mathrm{w}}) C^{\mathrm{f}}(\mathbf{x}_{\mathrm{w}}, t)}{M_0}, \tag{3}$$

where $f_{\mathrm{t}}(t; \mathbf{x} = \mathbf{x}_{\mathrm{w}}, \mathbf{x}_0)$ is travel time probability from the source at $\mathbf{x}_0$ to $\mathbf{x} = \mathbf{x}_{\mathrm{w}}$, $|q|$ is the local magnitude of the space- and time-varying specific discharge, $C^{\mathrm{f}}(\mathbf{x}_{\mathrm{w}}, t)$ is flux concentration, A is the area perpendicular to flow across which flux concentration is defined, and M_0 is the source mass. (Ref. [17] incorrectly neglects to put the absolute value on the specific discharge.) Rearranging (3) shows that

$$C^{\mathrm{f}}(\mathbf{x}_{\mathrm{w}}, t) = \frac{M_0}{|q(\mathbf{x}_{\mathrm{w}}, t)| A(\mathbf{x}_{\mathrm{w}})} f_{\mathrm{t}}(t; \mathbf{x}_{\mathrm{w}}, \mathbf{x}_0), \tag{4}$$

and by taking the derivative of C^{f} with respect to M_0, we obtain

$$\begin{aligned} f_{\mathrm{t}}(t; \mathbf{x}_{\mathrm{w}}, \mathbf{x}_0) &= |q(\mathbf{x}_{\mathrm{w}}, t)| A(\mathbf{x}_{\mathrm{w}}) \frac{\mathrm{d}C^{\mathrm{f}}(\mathbf{x}_{\mathrm{w}}, t)}{\mathrm{d}M_0} \\ &= |q(\mathbf{x}_{\mathrm{w}}, t)| A(\mathbf{x}_{\mathrm{w}}) \psi_{\mathrm{t}}(\mathbf{x}_{\mathrm{w}}, t; \mathbf{x}_0), \end{aligned} \tag{5}$$

where $\psi_{\mathrm{t}}(\mathbf{x}_{\mathrm{w}}, t; \mathbf{x}_0)$ is the state sensitivity of flux concentration at $\mathbf{x}_{\mathrm{w}}$, at any time t, to the source mass, M_0, at $\mathbf{x}_0$. Forward travel time probability is proportional to this sensitivity, and to the temporally-varying specific discharge at $\mathbf{x}_{\mathrm{w}}$. Note again that if there are multiple sources, each has its own forward travel time probability, an important concept for capture zone delineation.

These forward probabilities are related to sensitivities of concentration (C or C^{f}) at any location or time to the source mass at a known source location and release time; therefore, they are useful if the source is known. In the problem addressed here, however, the source is unknown and contamination is observed at one known location and time. We develop backward probabilities that describe either the possible source (or prior) locations of the observed contamination, with $\mathbf{x}_0$ as a random variable; or the release time of the contamination from a known source, with the release time as a random variable. These backward probabilities are related to the sensitivity of concentration at one *known* location and time to the source mass at any *random* upgradient location or *random* earlier time. These new sensitivities are, by definition, adjoint states [7], and their governing equation is the adjoint of a conventional contaminant transport equation (forward model). The adjoint equation models the same physical processes as the forward equation; however, the flow of information is reversed, so transport is modeled upgradient and backward in time from the observation to the possible sources or prior locations. Dispersion increases backward in time. Unlike the forward modeling approach, this adjoint (backward) model requires only one model simulation for each observation to obtain probabilities for all

possible sources, and therefore is more computationally efficient.

Backward probabilistic models have seen limited use in groundwater transport modeling through both random walk techniques and continuum methods. Uffink [22] and Chin and Chittaluru [2] used backward random walk methods to probabilistically delineate pumping well capture zones, and Fogg et al. [8] used it for groundwater vulnerability assessment. Bagtzoglou et al. [1] used a backward random walk to obtain probability maps to identify sources of contamination. Wilson and Liu [13,14,23] used a heuristically-developed backward continuum model to identify the source of groundwater contamination observed in a pumping well. They then tested this heuristic model using results of a field tracer test at the Canadian Forces Base, Borden, Ont., Canada [24]. The model results compared well with the tracer test data, especially for tracers that were injected near the extraction well; however, no formal justification was given for their model. Because their continuum model was developed heuristically, extending their approach to more complex systems is difficult.

Neupauer and Wilson [16,17] showed that the adjoint of a forward model is the appropriate continuum model for backward probabilities. This provided a rigorous mathematical approach for obtaining the backward model for any flow and transport system modeled with continuum equations. They developed the backward probability model for steady, uniform flow fields, and for steady non-uniform flow caused by spatial variability in hydraulic conductivity or by pumping. In the special case of constant porosity considered in [16,17], forward and backward location probabilities are equivalent for a specific source/observation pair and are an adjoint state of resident concentration; while forward and backward travel time probabilities are not equivalent to each other, and are related to, but not equivalent to, an adjoint state of resident concentration.

In this paper, we develop the continuum backward probability model for more general non-uniform flow and for transient flow. In particular, we address three important extensions: (1) spatially-varying porosity, (2) transient flow and temporally-varying porosity, and (3) distributed internal sources and sinks of water and solute. We illustrate these extensions using a vertically-integrated model, creating transient flow by a step change in pumping and non-uniform flow by a distributed internal source of areal recharge. The results demonstrate how the backward probability model can be applied to other, perhaps more important, non-uniform and transient flow conditions such as time-varying pumping or unsaturated (or saturated–unsaturated) flow and transport with spatially- and temporally-varying moisture content. Because the results are easier to compare in one dimension, we illustrate the approach for a one-dimensional, vertically-integrated aquifer; however, the relationships between adjoint states and probabilities are general to multidimensional systems. Multidimensional (2-D or 3-D) adjoint equations can be derived following the approach of [17]. Furthermore, although our forward contaminant transport model is a linear partial differential equation, the approach can also be applied to non-linear problems such as density-dependent flow. The method is applicable to a wide variety of physical, chemical, and biological transport processes that can be modeled as partial differential equations. In our work, we limit our analysis to systems that include, at a minimum, advection and dispersion.

We show that in general forward and backward location and travel time probabilities are not equivalent to adjoint states, but are related to them. Earlier work with constant porosity [16,17] did not encounter this generality, illustrating the importance of using a robust mathematical technique to develop the backward model when adding new complexities, whether using a continuum or random walk approach. While for each new situation the adjoint approach must be reapplied to determine the proper backward continuum model, the correct backward Fokker–Planck-type equation must also be rederived for each new backward random walk model. We also show that with recharge, or with any other distributed internal source of water, two different interpretations of backward probability can be made, depending on whether or not contamination enters through the source of water, or independent of it.

Although the state variable in the backward probability model is related to a probability density function, the model we present here is deterministic, with known parameter values. The adjoint approach, which we use here to develop the backward probability model, can also be applied in a general stochastic framework, addressing, for example, random permeability fields or scale-dependent dispersion [25].

2. Backward probability model

The governing equation for the backward probability model is the adjoint of the governing equation of forward contaminant transport. Transport of a conservative chemical in a one-dimensional flow field with spatially- and temporally-varying porosity can be modeled using the advection-dispersion equation (ADE), given by

$$\frac{\partial}{\partial t}(\theta C) = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(\theta v C) + q_{\mathrm{I}} C_{\mathrm{I}} - q_{\mathrm{O}} C, \qquad (6)$$

$$C(x,0) = C_{\mathrm{i}}(x),$$

$$\frac{\partial C}{\partial x} = g_1(t) \quad \text{at } x = x_1,$$

$$C(x,t) = g_2(t) \quad \text{at } x = x_2,$$

where C is resident concentration, t is time, x is the spatial direction, $D = a_L|v|$ is the dispersion coefficient, a_L is the longitudinal dispersivity, v is the groundwater velocity, θ is porosity, q_I is the source inflow rate per unit volume, C_I is the source strength, q_O is the sink outflow rate per unit volume, C_i is the initial concentration, x_1 and x_2 are the boundaries, and g_1 and g_2 are known functions. Other boundary conditions can also be used.

Location and travel time probabilities are related to sensitivities of concentration to the source mass (see (2) and (5)), and therefore can be obtained through sensitivity analysis. Neupauer and Wilson [16,17] previously used sensitivity analysis to obtain the backward model and probability relationships for the constant-porosity case; we also use that approach here but with non-constant porosity. The results for this new derivation are not an obvious extension of the previous work and lead to new results.

We begin by defining a performance measure, P, that quantifies some state of the system, defined as

$$P = \int\int_{x,t} h(\alpha, C)\,\mathrm{d}x\,\mathrm{d}t, \tag{7}$$

where $h(\alpha, C)$ is a functional of the state of the system, α is a system parameter, C is resident concentration, and integration is over the entire space–time domain. The marginal sensitivity of this performance measure with respect to the parameter α is obtained by differentiating (7) with respect to α:

$$\frac{\mathrm{d}P}{\mathrm{d}\alpha} = \int\int_{x,t} \left[\frac{\partial h(\alpha, C)}{\partial \alpha} + \frac{\partial h(\alpha, C)}{\partial C}\psi\right]\mathrm{d}x\,\mathrm{d}t, \tag{8}$$

where $\mathrm{d}P/\mathrm{d}\alpha$ is the marginal sensitivity, $\psi = \partial C/\partial\alpha$ is the state sensitivity, and $\partial h/\partial C$ is the Frèchet derivative [20] of the performance functional, h, with respect to C.

The choice of performance measure, P, depends on the type of probability that is desired. From (2), we see that location probability is related to the sensitivity of resident concentration at the observation point to the source mass; therefore, for location probability, the appropriate performance measure, P, is resident concentration at the observation point ($P = C(x_w, t = T)$) and the appropriate parameter α is the source mass ($\alpha = M_0$). Inspection of (7) shows that to obtain this performance measure for location probability, the performance functional must be defined as

$$h(x,\tau) = C\delta(x - x_w)\delta(\tau), \tag{9}$$

where $\tau = T - t$ is backward time, or time prior to sampling, and $\delta(\cdot)$ is a Dirac delta function.

Likewise, from (5), we see that backward travel time probability is related to the marginal sensitivity of flux concentration at the observation point to the source mass; therefore, for travel time probability, we define P to be flux concentration at the observation point ($P = C^f(x_w, t = T)$) and $\alpha = M_0$, and the performance functional, h, is defined as

$$h(x,\tau) = C^f\delta(x - x_w)\delta(\tau). \tag{10}$$

For both types of probability, $\alpha = M_0$ and (8) reduces to

$$\frac{\mathrm{d}P}{\mathrm{d}M_0} = \int\int_{x,t} \frac{\partial h}{\partial C}\psi\,\mathrm{d}x\,\mathrm{d}t, \tag{11}$$

where ψ is defined more explicitly now as $\psi = \partial C/\partial M_0$. Once the state sensitivity, ψ, is defined, we can use h from either (9) or (10) in this equation to obtain the marginal sensitivity; from this marginal sensitivity, we can calculate either location or travel time probability. The state sensitivity, ψ, can be obtained directly from (6) by differentiating each term with respect to M_0 to obtain a new form of the ADE in terms of the state sensitivity, $\psi = \partial C/\partial M_0$. The solution of this modified ADE produces the sensitivity of concentration at any location to the source mass, M_0, at one location. This result is useful if the source is known; however, if contamination is observed in the aquifer and the source of contamination is unknown, we are more interested in the sensitivity of concentration at a *one* location (e.g., at x_w, the location of a monitoring or pumping well) to the source mass at *any* location. Therefore, we would need to calculate $\psi(x_w, t)$ for many different source locations, which could be obtained rather inefficiently through repeated simulations of (6) with different x_0. The number of simulations needed would depend on the number of possible source locations and on the spatial discretization of the problem domain. As a worst case scenario, if no information about the source location was available, then (6) would have to be run once for each cell in the domain, with the source location in a different cell for each simulation. Also, to identify the travel time with no information on the source release time, (6) would have to be solved once for each time step.

Instead, we take a more efficient approach of replacing (11) with an equivalent expression that does not contain ψ, but contains an arbitrary adjoint state, ψ^*, which we later show to be equivalent to the desired sensitivity (i.e., the sensitivity of concentration at *one* location to the source mass at *any* location). With this replacement, the marginal sensitivity becomes (see Appendix A)

$$\frac{\mathrm{d}P}{\mathrm{d}M_0} = \int_x (\theta\psi^*)\Big|_{t=0} \frac{\partial C_i}{\partial M_0}\mathrm{d}x, \tag{12}$$

where $C_i = C(x, 0)$ is the initial concentration in (6), and the adjoint state, ψ^*, is the solution to the adjoint equation (see Appendix A)

$$\frac{\partial}{\partial\tau}(\theta\psi^*) = \frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi^*}{\partial x}\right) + \frac{\partial}{\partial x}(\theta v\psi^*) - q_1\psi^* + \frac{\partial h}{\partial C}, \tag{13}$$

$$\psi^*(x,0) = 0,$$

$$D\frac{\partial\psi^*}{\partial x} + v\psi^* = 0 \quad \text{at } x = x_1,$$

$$\psi^* = 0 \quad \text{at } x = x_2,$$

where τ is backward time representing time prior to observation, the load term, $\partial h/\partial C$, is the same Frèchet derivative as in (11), and the performance functional, h, is defined in (9) and (10) for location and travel time probabilities, respectively. Since backward time τ is the time prior to observation, it increases as forward time t decreases; thus the time derivatives in this equation and (6) have opposite signs, allowing for the flow of information in reversed time in (13). The other differences between the forward equation (6) and the backward (adjoint) equation (13) are the reversal of sign on the velocity term allowing upgradient flow of information and the replacement of sources in the forward model with sinks in the backward model as a consequence of the flow-field reversal. The sink of solute in the forward model does not become a source in the backward model. These differences cause information to be propagated from the observation back to all possible prior locations. In other words, with just one simulation of (13), we obtain information about all possible source locations for the observed contamination. Recall that obtaining the same information from (6) would require one forward simulation for each possible source. It is also important to observe that the dispersion term in (13) causes spreading of the adjoint states, and therefore of the backward probabilities, backward in time.

The boundary conditions in the backward model are different than those of the forward model. In the adjoint model, all boundary conditions are homogeneous. The first-type boundaries in the forward model remain first-type in the backward model; however, the second-type boundaries in the forward model become third-type in the backward model, and vice versa [17]. Both (12) and (13) differ from the equations in [17] by the explicit inclusion of the spatial and temporal dependence of porosity.

At this point, we have an efficient approach for calculating marginal sensitivities. We calculate the adjoint state by substituting the performance functional, h, into the load term in (13), and then we use the results in (12) to calculate the marginal sensitivity. The remaining task is to relate the marginal sensitivity to the probability of interest. We show how the marginal sensitivities are related to both forward and backward probabilities.

For location probability, the performance functional h is (9), leading to $\partial h/\partial C = \delta(x - x_w)\delta(\tau)$ as the load term in the adjoint equation. Let ψ_x^* denote the solution to (13) with this load term and let $C_i = [M_0/\theta(x_0, t = 0)]\delta(x - x_0)$ in (12) for an instantaneous point source of contamination. The marginal sensitivity from (12) reduces to

$$\frac{dC(x_w, \tau = 0)}{dM_0(x_0)} = \psi_x^*(x_0, \tau = T; x_w), \tag{14}$$

where x_w and T are fixed and x_0 is random. This adjoint state, ψ_x^*, describes the sensitivity of the observed concentration at x_w at $\tau = 0$ to a release of mass at any prior location, x_0, at backward time $\tau = T$. Contrast this with the state sensitivity ψ_x in (2) that describes the sensitivity of concentration at any location and time to a source mass at a specific x_0 and at time $t = 0$.

Both sensitivities (ψ_x and ψ_x^*) are related to forward location probability, each with a different point of view. From (2), forward location probability at x_w due to a source at x_0 is

$$\begin{aligned} f_x(x = x_w; t = T, x_0) &= \theta(x_w, t = T)\frac{dC(x_w, t = T; x_0)}{dM_0(x_0)} \\ &= \theta(x_w, t = T)\psi_x(x_w, t = T; x_0), \end{aligned} \tag{15}$$

representing the probability that contamination from x_0 will be at a random x_w at time $t = T$. If we have one source and many possible observers, it is efficient to calculate forward location probability using (15) with ψ_x, the sensitivity of concentration at any location to a source mass at a known source, since ψ_x can be calculated from one simulation of the forward model. If we have many possible sources and one observer, it is more efficient to use ψ_x^*, the sensitivity of concentration at one location to a source mass anywhere; ψ_x^* can be calculated from one simulation of the backward model (13). Using (14) in (15), the forward location probability is then given by

$$f_x(x = x_w; t = T, x_0) = \theta(x_w, \tau = 0)\psi_x^*(x_0, \tau = T; x_w). \tag{16}$$

Note that the adjoint state and porosity are defined in terms of backward time τ, while the forward location probability is defined in terms of forward time t.

The adjoint state ψ_x^* is also related to backward location probability by

$$f_x(x = x_0; \tau = T, x_w) = \theta(x_0, \tau = T)\psi_x^*(x_0, \tau = T; x_w), \tag{17}$$

where $f_x(x = x_0; \tau = T, x_w)$ represents the probability that contamination observed at x_w was at x_0 (random variable) at time T in the past. Note the subtle differences between the expressions for forward and backward probabilities in (16) and (17). They are functions of the same adjoint state, but they depend on the "local" value of porosity. Forward probability (16) is a function of the porosity at the observer (prediction location x_w and time $t = T$), and backward probability (17) is a

function of the porosity at the "source" or prior location (at x_0 and time $\tau = T$). A smaller local porosity indicates a smaller pore volume and thus a lower probability of finding the solute parcel in the same bulk volume of aquifer material. If porosity is uniform, the two location probabilities are equivalent [17].

Next we show how travel time probability can be obtained from (12) and (13). For travel time probability, the performance functional h is (10). Since the load term in the adjoint equation (13) is the Frèchet derivative of h with respect to C, but (10) is written in terms of C^{f}, we must substitute into (10) the relationship between C and C^{f}. In one dimension, this relationship is [18]

$$C^{\mathrm{f}} = C - \frac{D}{v}\frac{\partial C}{\partial x}. \tag{18}$$

Substituting this expression into (10) and taking the Frèchet derivative with respect to C, we obtain the load term for backward travel time probability, given by

$$\frac{\partial h}{\partial C} = \delta(x - x_{\mathrm{w}})\delta(\tau) + \frac{D}{v}\delta'(x - x_{\mathrm{w}})\delta(\tau), \tag{19}$$

where $\delta'(x)$ is the derivative of the Dirac delta function with respect to x [16]. We use this load term in (13) for travel time probability for contamination observed at a monitoring well, but we use a different load term for contamination observed at a pumping well. At a pumping well, we often assume that $\partial C/\partial x = 0$; therefore from (18), $C^{\mathrm{f}} = C$ at the well, and the load term in (13) is

$$\frac{\partial h}{\partial C} = \delta(x - x_{\mathrm{w}})\delta(\tau). \tag{20}$$

This is the same load term used for location probability.

Let ψ^*_τ be the solution to (13) with either (19) or (20) as the load term, and let $C_{\mathrm{i}} = [M_0/\theta(x_0, t = 0)]\delta(x - x_0)$ in (12). The marginal sensitivity in (12) reduces to

$$\frac{\mathrm{d}C^{\mathrm{f}}(x_{\mathrm{w}}, \tau = 0; x_0)}{\mathrm{d}M_0(x_0)} = \psi^*_\tau(x_0, \tau = T; x_{\mathrm{w}}). \tag{21}$$

The adjoint state, ψ^*_τ, describes the sensitivity of flux concentration at a specific x_{w} to a release of mass at any prior location, x_0. Contrast this with the state sensitivity ψ_{t} in (5) that describes the sensitivity of flux concentration at any x_{w} to a release of mass at a specific location x_0.

Both ψ^*_τ and ψ_{t} are related to forward travel time probability, which represents the probability that contamination from x_0 will arrive at x_{w} at time t (random variable) in the future. From (5), forward travel time probability is related to the sensitivity, $\mathrm{d}C^{\mathrm{f}}/\mathrm{d}M_0$. Since this sensitivity can be represented by ψ^*_τ or ψ_{t} (see (5) and (21)), travel time probability can also be described using either ψ^*_τ or ψ_{t}. If we have one source and many possible observers, ψ_{t} can be obtained with one simulation of a forward model, and forward travel time probability can be obtained efficiently using ψ_{t}. On the other hand, if we have one observer and many possible sources x_0, it is more efficient to use ψ^*_τ to obtain forward travel time probability because ψ^*_τ can be obtained with one simulation of a backward model. From (5) and (21), these expressions for forward travel time probability are

$$\begin{aligned} f_{\mathrm{t}}(t = T; x_{\mathrm{w}}, x_0) &= |q(x_{\mathrm{w}}, t = T)|A(x_{\mathrm{w}})\psi_{\mathrm{t}}(x_{\mathrm{w}}, t = T; x_0) \\ &= |q(x_{\mathrm{w}}, \tau = 0)|A(x_{\mathrm{w}})\psi^*_\tau(x_0, \tau = T; x_{\mathrm{w}}). \end{aligned} \tag{22}$$

Recall that the first expression in (22) is used when the source is known and there are many possible receptors; while the second expression can be used if there are many possible sources x_0 (or prior locations), each with its own possible travel time T.

Note that $\int_0^t f_{\mathrm{t}}(t'; x_{\mathrm{w}}, x_0)\,\mathrm{d}t'$ represents the probability that contamination from a source at x_0 will reach x_{w} in time t or less. In [17], Neupauer and Wilson show that this integral of forward travel time probability can be used to delineate a probabilistic capture zone for a pumping well. In this type of application, we have one observer (the pumping well) and many possible sources; therefore, forward travel time probability capture zones can be obtained efficiently using ψ^*_τ.

The adjoint state ψ^*_τ is also related to backward travel time probability by

$$f_\tau(\tau = T; x_0, x_{\mathrm{w}}) = |q(x_0, \tau = T)|A(x_0)\psi^*_\tau(x_0, \tau = T; x_{\mathrm{w}}), \tag{23}$$

in a one-dimensional domain, where $f_\tau(\tau = T; x_0, x_{\mathrm{w}})$, represents the probability that contamination observed at x_{w} was at x_0 at a random time $\tau = T$ in the past. Again note the subtle differences between the expressions for forward (22) and backward (23) probabilities when expressed in terms of the adjoint state ψ^*_τ. They depend on the local rate of groundwater flow. Where or when the flow is slower, the travel time probability decreases.

In the special case of constant porosity, the ADE (6) reduces to

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(vC) + \frac{q_{\mathrm{I}}}{\theta}C_{\mathrm{I}} - \frac{q_{\mathrm{O}}}{\theta}C, \tag{24}$$

and the adjoint equation is [17]

$$\frac{\partial \psi^{**}}{\partial \tau} = \frac{\partial}{\partial x}\left(D\frac{\partial \psi^{**}}{\partial x}\right) + \frac{\partial}{\partial x}(\psi^{**}C) - \frac{q_{\mathrm{I}}}{\theta}\psi^{**} + \frac{\partial h}{\partial C}, \tag{25}$$

with the marginal sensitivity defined as [17]

$$\frac{\mathrm{d}P}{\mathrm{d}M_0} = \int_x \psi^{**}\bigg|_{t=0}\frac{\partial C_{\mathrm{i}}}{\partial M_0}\,\mathrm{d}x, \tag{26}$$

where the relationship between the adjoint state ψ^{**} in this equation and the adjoint state ψ^* in the rest of this paper is $\psi^{**} = \theta\psi^*$. With $C_{\mathrm{i}} = (M_0/\theta)\delta(x - x_0)$, (26) simplifies to $\mathrm{d}C/\mathrm{d}M_0 = \psi^{**}_x/\theta$ for location probability, and from (17), $f_x = \psi^{**}_x$ for both forward and backward

location probabilities, explaining their reciprocal relationship. Neupauer and Wilson [17] obtained this result directly using $\alpha = M' = M_0/\theta$.

The adjoint derivation presented here is easily extended to multiple dimensions [17]. The relationships between adjoint states and probabilities in (16), (17), (22), and (23) are valid in multiple dimensions by replacing the scalar location x with a location vector.

3. Illustrations of the backward probability model

In this section we present several illustrations of the backward probability model for a conservative tracer using the hypothetical vertically-integrated aquifer shown in Fig. 1. The confined aquifer is semi-infinite and one-dimensional with a pumping well at $x_w = 0$, and an instantaneous point source of contamination at $x_0 > 0$. Initially, we assume uniform porosity and steady flow (from right to left in the figure), with no internal sources or sinks of water or contamination. Later we relax these assumptions, to encompass the three extensions of previous work, including (1) spatially-varying porosity, (2) transient flow and temporally-varying porosity, and (3) distributed internal sources and sinks of water and solute.

3.1. Backward probability model in a steady and uniform flow field

The governing equation for the forward model is (6), with $q_I = q_O = 0$, $x_1 = 0$, $x_2 \to \infty$, $g_1(t) = g_2(t) = 0$, and $C_i(x) = (M_0/\theta)\delta(x - x_0)$. The solution to (6) for constant v, D, and θ is [23]

$$C(x,t) = \frac{M_0}{\theta\sqrt{4\pi Dt}} \exp\left\{-\frac{(x - x_0 - vt)^2}{4Dt}\right\} \times \left[1 + \exp\left\{\frac{-x_0 x}{Dt}\right\}\right] + \frac{M_0 v}{2D\theta} \times \exp\left\{\frac{-vx_0}{D}\right\}\mathrm{erfc}\left[\frac{x + x_0 - vt}{\sqrt{4Dt}}\right], \quad (27)$$

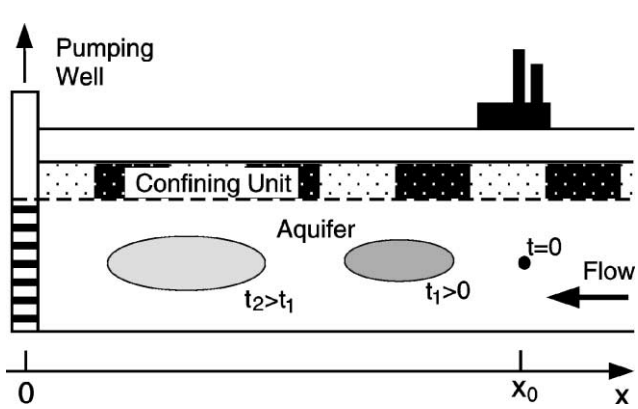

Fig. 1. Aquifer geometry for example problem.

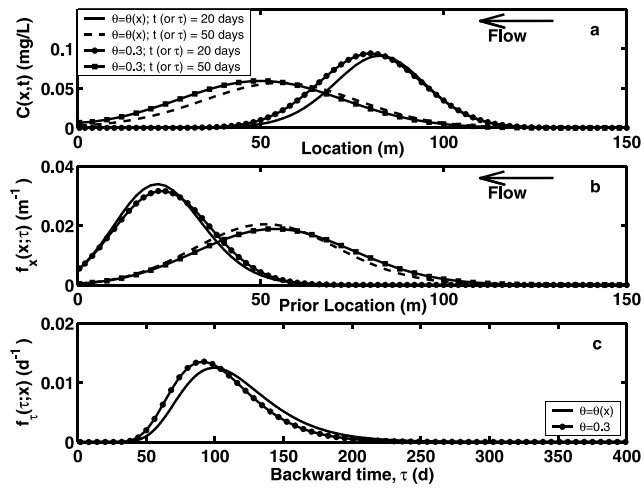

Fig. 2. Results of simulations with constant porosity and with spatially-varying porosity ($v = -1$ m/d at x_w, $a_L = 5$ m). (a) Resident concentration ($M_0 = 1$ g/m^2; $x_0 = 100$ m). (b) Backward (prior) location probability ($x_w = 0$). (c) Backward travel time probability ($x_w = 0$, $x_0 = 100$ m).

Table 1
Transport parameters for the example problem

Parameter	Value
Source location, x_0 (m)	100
Groundwater velocity, v (m/d)	−1
Longitudinal dispersivity, a_L (m)	5
Porosity, θ	0.3
Source mass, M_0 (g/m^2)	1

for $v < 0$ (i.e., flow in the direction of $-x$). This solution is plotted in Fig. 2a showing resident concentration for $t = 20$ days (circles) and $t = 50$ days (squares) after release from the source at $x_0 = 100$ m using the transport parameters values in Table 1. At $t = 20$ days, the contamination has not yet reached the pumping well, but by $t = 50$ days, some contamination has reached the well.

Suppose one sample of contamination is observed at the pumping well at time $\tau = 0$, and we want to know its location at some time in the past. To determine likely prior locations, we use backward location probability. The governing equation for this case is (13) with h defined in (9), $q_I = 0$, $x_1 = 0$, and $x_2 \to \infty$. The solution to this equation, ψ_x^*, is used in (17) to obtain backward location probability, given by [23]

$$f_x(x;\tau) = \frac{1}{\sqrt{\pi D\tau}} \exp\left\{-\frac{(x + v\tau)^2}{4D\tau}\right\} + \frac{v}{2D}\exp\left\{\frac{-vx}{D}\right\}\mathrm{erfc}\left[\frac{x - v\tau}{\sqrt{4D\tau}}\right], \quad (28)$$

for $v < 0$. Backward location probability is plotted in Fig. 2b for $\tau = 20$ days (circles) and $\tau = 50$ days (squares) prior to observation at the pumping well,

showing the probable positions of the observed contamination at $\tau = 20$ and $\tau = 50$ days prior to sampling. The most likely prior position of the contamination is $x \approx 23$ m at $\tau = 20$ days prior to sampling, and $x \approx 55$ m at $\tau = 50$ days prior to sampling. Note also that f_x ($x = 100$ m; $\tau = 20$ days) ≈ 0; this is consistent with Fig. 2a (circles), which shows that mass originating at $x_0 = 100$ m does not reach the pumping well in 20 days. At $\tau = 50$ days, however, the location probability at $x = 100$ m is non-zero, indicating that the mass that originated at $x_0 = 100$ m has a finite probability of reaching the pumping well in 50 days. This is also consistent with Fig. 2a (squares).

Suppose, instead, that we would like to know when the observed contamination could have been released from a known or suspected source at $x = x_0 = 100$ m. To determine the likely travel times from the source to the pumping well, we use backward travel time probability. The governing equation for backward travel time probability is (13) with the load term defined in (20) for observation at a pumping well; this is the same as the governing equation for location probability, so $\psi^*_\tau = \psi^*_x$ for this special case.

From (23) and (28), backward travel time probability for this case is

$$f_\tau(\tau; x) = \frac{-v}{\sqrt{\pi D \tau}} \exp\left\{ -\frac{(x + v\tau)^2}{4D\tau} \right\} - \frac{v^2}{2D} \exp\left\{ \frac{-vx}{D} \right\} \operatorname{erfc}\left[\frac{x - v\tau}{\sqrt{4D\tau}} \right], \tag{29}$$

where $|q|$ in (23) is $|q| = -v\theta$ because $v < 0$ in this example. This travel time probability distribution, plotted in Fig. 2c (circles), shows that the most likely travel time from $x_0 = 100$ m to the pumping well is $\tau \approx 92$ days. Note also that $f_\tau > 0$ at $\tau = 50$ days, indicating a non-zero probability that mass from $x_0 = 100$ m will reach the pumping well in 50 days. This is consistent with the results shown in Fig. 2a (squares and circles).

3.2. Backward probability model with spatially-varying aquifer porosity

The next illustration addresses spatial variability in aquifer porosity which enters both the adjoint equation and the relationships between adjoint states and probabilities in (17) and (23). To simplify the comparison of results, we use a linear variation in porosity, given by

$$\theta(x) = \theta_0 + 0.0005x, \tag{30}$$

where $\theta_0 = 0.3$ is the porosity at $x = 0$ and x has units of m. The same approach would be followed for other spatial distributions, including realizations of random fields.

For a one-dimensional domain with no internal sources or sinks of water, as in Fig. 1, the specific discharge, q, is uniform because of mass conservation. If aquifer porosity is spatially-varying, groundwater velocity and the dispersion coefficient vary in space, because $v(x) = q/\theta(x)$, and the governing equations (6) and (13) must be solved numerically. We used MOD FLOW-96 [9] and MT3DMS [26] to simulate steady flow and transient transport using the parameter values shown in Tables 1 and 2. With these values and (30), the velocity at the well (at $x = 0$) is $v = -1$ m/d, and its magnitude decreases upgradient of the well.

Table 2
Flow parameters for numerical simulations

Parameter	Value
Downstream boundary, x_1	0
Upstream boundary, x_2 (m)	201
Specified head at x_2 (m)	100
Spatial discretization (m)	2 (1 m at x_1)
Aquifer thickness, B (m)	10
Aquifer width (m)	1
Transmissivity (m^2/s)	0.0002
Specific storage, S_s (m^{-1})	0.001
Pumping rate, Q (m^3/d)	3

Fig. 2a shows the resident concentration in the variable-porosity aquifer at $t = 20$ days (solid line) and $t = 50$ days (dashed line) after release from the source at $x_0 = 100$ m. For comparison, the plumes for the constant porosity ($\theta = 0.3$) case are also shown. With spatially-varying porosity, the porosity at the source (at $x_0 = 100$ m) is $\theta(x_0) = 0.35$; therefore the velocity ($v = q/\theta$) near the source is slower for the spatially-varying porosity case. This is confirmed in Fig. 2a, which illustrates that the constant-porosity plume moves faster than the spatially-varying-porosity plume (compare circles and solid line). Also, because porosity is higher in the aquifer with spatially-varying porosity, the spatially-varying-porosity plume occupies a smaller portion of the total aquifer volume.

If contamination is observed in the pumping well, we can use backward location probability to determine where the contamination was at some time in the past. The appropriate governing equation is (13) with h as defined in (9). We solved this equation numerically for ψ^*_x using MODFLOW-96 and MT3DMS with the parameters in Tables 1 and 2 and porosity in (30) (see [15] for details of the numerical implementation of the backward probability model), and we used the results in (17) to calculate backward location probability. Fig. 2b shows backward location probability for $\tau = 20$ days (solid line) and $\tau = 50$ days (dashed line) prior to observation at the pumping well. For comparison, the location probability for the constant-porosity case is also shown. Because the velocity is slower for the spatially-varying-porosity case, the possible prior locations are slightly closer to the well, and the distribution is less disperse.

Using $\psi^*_\tau = \psi^*_x$ in (23), we also calculated backward travel time probability from the pumping well to the source at $x = x_0 = 100$ m. For contamination that is observed in the pumping well, the backward travel time probability plotted in Fig. 2c shows the possible times in the past that the contamination was at $x_0 = 100$ m (solid line). The travel time probability for the constant-porosity case is also shown in the figure (circles). Because the velocity is slower for the spatially-varying-porosity case, the travel time probability distribution is shifted to later backward times, suggesting an earlier actual release time at the source.

Although this illustration implements the backward probability model for spatially-varying porosity, it is a surrogate for implementation in similar situations with spatially-varying storage properties. Examples include variable saturated thickness in a vertically integrated Dupuit model of a phreatic aquifer (or the upper layer of a 3D phreatic aquifer application of MODFLOW-96 and MT3DMS), and unsaturated flow with spatially-varying moisture content. In general, spatial variability of porosity, saturated thickness, or moisture content is incorporated into the governing equation (13), which is solved for adjoint states in the usual manner. The backward probabilities are further affected by the spatial variability because porosity appears in the relationships between the adjoint states and probabilities, (17) and (23).

3.3. Backward probability model with transient flow

The next illustration addresses transient flow and temporal variability of porosity. We have chosen to vary the velocity through a step change in the pumping rate at the downgradient domain boundary. Neglecting water compressibility, the accumulation term of the continuity equation becomes $S_s(\partial h/\partial t) = \partial\theta/\partial t$, where S_s is specific storage and h is hydraulic head, indicating that porosity is also temporally variable. This example, therefore, is a surrogate for any process that produces quantifiable temporal variability in storage properties, including saturated thickness in vertically-integrated models and moisture content in unsaturated flow.

If the flow field in an aquifer is transient, the forward contaminant transport model is governed by (6), with $v = v(x,t)$. For emphasis, we generalize (6) for the temporally-varying parameters as

$$\frac{\partial}{\partial t}[\theta(x,t)C] = \frac{\partial}{\partial x}\left[\theta(x,t)D(x,t)\frac{\partial C}{\partial x}\right] - \frac{\partial}{\partial x}[\theta(x,t)v(x,t)C] + q_I C_I - q_O C. \quad (31)$$

The governing equation for the backward probability model is the adjoint of (31), rewritten here to emphasize the temporal and spatial variability in the parameter values:

$$\frac{\partial}{\partial \tau}[\theta(x,T-\tau)\psi^*] = \frac{\partial}{\partial x}\left[\theta(x,T-\tau)D(x,T-\tau)\frac{\partial \psi^*}{\partial x}\right] + \frac{\partial}{\partial x}[\theta(x,T-\tau)v(x,T-\tau)\psi^*] - q_I\psi^* + \frac{\partial h}{\partial C}, \quad (32)$$

where we have substituted $t = T - \tau$. Note that the flow field in the backward (adjoint) model is reversed in both space (positive sign on the advection term) and time (now in terms of $T - \tau$) relative to the forward model, leading to a reversal in time of θ and D also. In other words, in the backward model, the transient behavior of θ and D must begin with its final state at $t = T$ ($\tau = 0$) and progress back to its original state at $t = 0$ ($\tau = T$). The backward location and travel time probabilities are still defined by (17) and (23), respectively.

Suppose the aquifer in Fig. 1 is initially at steady state with a pumping rate of $Q = 4$ m^3/d, and at $t = 0$ the pumping rate is decreased to $Q = 3$ m^3/d. After the pumping rate is decreased, head increases and the flow field is transient until a new steady state is reached. We used MODFLOW-96 and MT3DMS to solve (31) and (32) for this scenario. In MT3DMS, porosity is not updated during the simulation (i.e., $\theta \neq \theta(t)$), therefore we only considered temporally-varying v and D and were not able to simulate temporally-varying porosity (see Appendix B for more details).

The head distribution in the aquifer at five different times is shown in Fig. 3. The aquifer reaches a new steady state in approximately 40 days. Fig. 4a shows resident concentration at $t = 20$ days (solid line) and $t = 50$ days (dashed line) after release from a source at $x_0 = 100$ m. For comparison, the plumes from the steady flow simulation ($Q = 3$ m^3/d; $v = -1$ m/d) are also shown. In the transient flow case, the velocity is initially faster than in the steady flow simulation; therefore, the plume moves slightly faster.

To calculate backward location probability, we solved (32) with h as defined in (9) for the adjoint state ψ^*_x using MODFLOW-96 and MT3DMS, with $\tau = T - t$ and $T = 50$ days. We used the results in (17) to calculate backward location probability. Fig. 4b shows the backward location probability distribution for $\tau = 20$ days (solid line) and $\tau = 50$ days (dashed line) prior to observation at the pumping well (at $x_w = 0$). For comparison, the location probability for the steady flow case

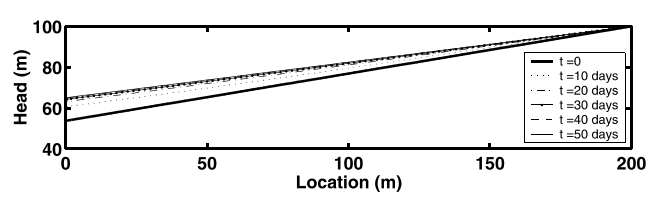

Fig. 3. Head distribution in transient flow simulation.

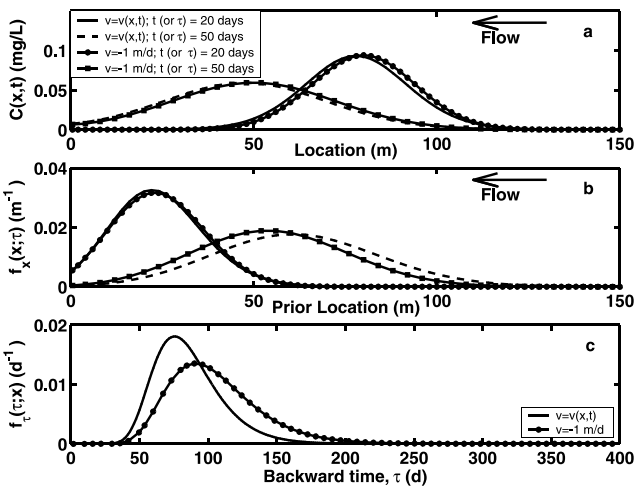

Fig. 4. Results of transient flow simulations ($v_0 = -1$ m/d, $a_L = 5$ m). (a) Resident concentration ($M_0 = 1$ g/m^2; $x_0 = 100$ m). (b) Backward (prior) location probability ($x_w = 0$). (c) Backward travel time probability ($x_w = 0$, $x_0 = 100$ m).

is also shown (circles and squares). For early τ (later forward time), the aquifer is essentially at steady state with the lower pumping rate ($Q = 3$ m^3/d; $v = -1$ m/d); therefore, the steady-flow and transient-flow location probability distributions at $\tau = 20$ days are essentially equivalent. At $\tau = 50$ days, the aquifer is at steady state with the higher pumping rate ($Q = 4$ m^3/d); therefore the transient-flow probability distribution is shifted farther upgradient than the steady-flow distribution.

The backward travel time probability from the pumping well to the source at $x = x_0 = 100$ m was calculated using $\psi_\tau^* = \psi_x^*$ in (23). These results are plotted in Fig. 4c (solid line), along with the travel time probability for the steady-flow case (circles). Because the velocity is faster in the transient-flow case, the travel time probability distribution is shifted to earlier backward times, indicating that a more recent release from the source is likely.

This example illustrates the implementation of the backward probability model with temporally-varying parameters values, cases such as unsaturated flow with temporally-varying moisture content. The temporal variability of parameters is incorporated into the governing equation (13), with the time-dependence of the parameters reversed to begin with the final state at $t = T$ ($\tau = 0$) and end with the original state at $t = 0$ ($\tau = T$).

3.4. Backward probability model with a distributed source

The final illustration addresses distributed internal sources or sinks of water and solute. We use natural recharge, a spatially-distributed source of water, and possibly of contamination, in a vertically-integrated aquifer model. In the absence of any other internal sources or sinks, the one-dimensional forward governing equation (6) is modified as

$$\frac{\partial(\theta C)}{\partial t} = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(\theta v C) + \frac{N}{B} C_I, \tag{33}$$

where the sink term in (6), $-q_O C$, is eliminated because we are assuming no internal sinks, N is the natural recharge rate (here assumed to be spatially-uniform), B is the aquifer thickness, and the ratio N/B is the source inflow rate, q_I. Assuming that the natural recharge is free of contamination (i.e., $C_I = 0$), the final term in (33) vanishes.

Since natural recharge is a spatially-distributed source of water, the velocity in the aquifer varies in space. With uniform natural recharge, the velocity in the aquifer shown in Fig. 1 is

$$v(x) = v_0 + \frac{Nx}{B\theta}, \tag{34}$$

where $v_0 = -1$ m/d is the velocity at the pumping well, and velocity decreases in magnitude upgradient of the well. We solved (33) numerically using MODFLOW-96 and MT3DMS with the parameters listed in Tables 1 and 2, and with $N = 0.009$ m/d. We used an unrealistically large N to amplify the effects of natural recharge. The results are plotted in Fig. 5a, showing the resident concentration for $t = 20$ days (solid line) and $t = 50$ days (dashed line) after release from the source. For comparison, the plumes for the no-recharge case are also shown. The plume with recharge travels more slowly and is less disperse than the plume without recharge because the magnitude of the velocity is lower.

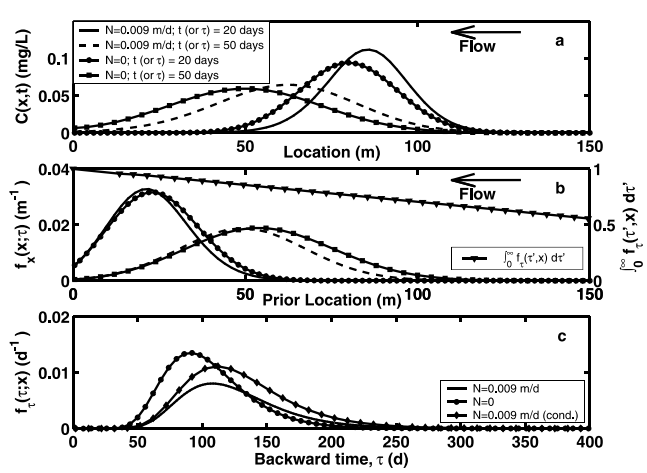

Fig. 5. Results of simulations with natural recharge ($v_0 = -1$ m/d, $a_L = 5$ m, $\theta = 0.3$). (a) Resident concentration ($M_0 = 1$ g/m^2; $x_0 = 100$ m). (b) Backward (prior) location probability and the integral over time of backward travel time probability, $f_\tau(\tau, x)$ (solid line with triangles) ($x_w = 0$). (c) Backward travel time probability ($x_w = 0$, $x_0 = 100$ m).

To calculate backward location probability with recharge, we used MODFLOW-96 and MT3DMS to solve for the adjoint state ψ_x^* in (13) with $q_I = N/B$, and we used the results in (17) to calculate location probability. Fig. 5b shows the backward location probability distribution for $\tau = 20$ days (solid line) and $\tau = 50$ days (dashed line) prior to observation at the pumping well (at $x_w = 0$). For comparison, the location probability for the no-recharge case is also shown. Because of a lower velocity, the location probability distribution with recharge is shifted toward the well (compare solid line and circles), indicating that contamination observed in the pumping well was likely to have been closer to the well at some time in the past.

The recharge term in the adjoint equation (13) has the same form as a first-order decay term with an equivalent decay rate of $\lambda = N/(B\theta) = 0.003\ \mathrm{d}^{-1}$ for the parameters used in this example. As can be seen in Fig. 5b, probability is "decaying", i.e., $\int_x f_x \,\mathrm{d}x < 1$, for $\tau > 0$, indicating a finite probability that the observed contamination entered the system through the natural recharge and therefore was not in the system at the time of interest. In other words, $1 - \int_x f_x(x;\hat{\tau})\,\mathrm{d}x$ is equivalent to the probability that the observed contamination entered the system via natural recharge during the time interval $0 < \tau < \hat{\tau}$. With recharge behaving like a first-order decay process, $\int_x f_x(x;\tau)\,\mathrm{d}x = \mathrm{e}^{-\lambda\tau}$, where $\lambda = N/(B\theta)$. For $\tau = 20$ days (solid line in Fig. 5b), $\int_x f_x(x;\tau)\,\mathrm{d}x = 0.9401$, and $\int_x f_x(x;\tau)\,\mathrm{d}x = 0.8622$ for $\tau = 50$ (dashed line in Fig. 5b). These values are essentially equivalent to $\mathrm{e}^{-\lambda\tau}$.

Suppose you know that the observed contamination did not enter the aquifer through recharge, i.e., $C_I = 0$ as we assumed in this example. In this case, there is a probability of zero that the contamination entered through natural recharge, requiring that $\int_x f_x(x;\tau)\ \mathrm{d}x = 1$ when integrated over the entire spatial domain. This second interpretation of backward location probability can be achieved by setting $q_I = 0$ in the adjoint equation (13). The backward probabilities are still influenced by recharge through the flow field, but they do not "decay" over time. Results for this case are not shown in Fig. 5b; however, they would be equivalent to normalizing the curves by $\int_x f_x(x;\tau)\,\mathrm{d}x = \mathrm{e}^{-\lambda\tau}$.

Backward travel time probability with recharge also has two interpretations. We first consider the scenario in which the contamination could have entered through natural recharge. Since $\psi_\tau^* = \psi_x^*$ for an observation at a pumping well, we use the adjoint state from the location probability simulation in (23) to calculate backward travel time probability. The results are plotted in Fig. 5c, showing the backward travel time probability from the pumping well to the source at $x = x_0 = 100$ m (solid line). For comparison, the travel time probability is also shown for the no-recharge case (circles). Because the velocity is slower with recharge, the travel time probability distribution is shifted toward later backward times, indicating an earlier actual release time at $x_0 = 100$ m. By comparing the areas under the curves in Fig. 5c, we see that travel time probability decays over time because the recharge term in the adjoint equation acts like a first-order decay term. The amount of decay, $1 - \int_\tau f_\tau(\tau;\hat{x},x_w)\,\mathrm{d}\tau$, represents the probability that the observed contamination entered the aquifer downgradient of $\hat{x}$ and therefore never was at $\hat{x}$. From Fig. 5c, $\int_0^\infty f_\tau(\tau',x_0)\,\mathrm{d}\tau' \approx 0.71$ at $x_0 = 100$ m, indicating a 71% probability that the observed parcel was ever at $x_0 = 100$ m, and a 29% probability that the parcel entered the system through natural recharge between $x_w = 0$ and $x_0 = 100$ m, and therefore was never at $x_0 = 100$ m. The values of this integral for other x are shown in Fig. 5b (triangles, right-hand axis). The probability that the observed contamination was ever at a location x decreases as the distance from the well increases.

If we know that the contamination did not enter through natural recharge, we could set $q_I = 0$ in (13), preventing the loss of backward probability through reversed natural recharge. This conditional travel time probability is shown in Fig. 5c (diamonds). The travel time probability is still shifted to later times relative to the no-recharge case; however, $\int_0^\infty f_\tau(\tau',x_0)\,\mathrm{d}\tau' = 1$, indicating that the observed contamination was at $x_0 = 100$ m at some time in the past and did not enter through natural recharge.

If natural recharge, or any other distributed internal source of water, is modeled, the user must decide which of the two interpretations is more appropriate. In some situations, natural recharge is a known pathway for contamination to enter the groundwater, so the first interpretation is more appropriate. In situations in which the contamination source is likely to be vertically distributed within the aquifer, the second interpretation is more appropriate.

This example illustrates the implementation of the backward probability model with internal sources or sinks of solute and water. Internal sources of solute become sinks of probability in the backward model, producing a "decaying" probability distribution. The probability integrates to less than unity over the appropriate domain (space for location probability; time for travel time probability), with the difference indicating the probability that the contamination entered through the internal source. For an internal source of water that does not contain the solute, the associated sink term can be eliminated from the backward probability model because the solute could not have entered there. A solute sink in the forward model does not appear in the backward model. The solute parcel that was found at the observation point obviously did not leave the system at the internal sink; therefore, the solute sink has no effect on the prior position of the solute parcel, except through its influence on the velocity field.

4. Conclusions

Backward location and travel time probabilities were developed heuristically by Liu [13] and Wilson and Liu [23] for several cases including transport of conservative solutes in uniform and non-uniform flow fields. Neupauer and Wilson [16,17] demonstrated that these backward probabilities are related to adjoint states of resident concentration, and they used adjoint theory to obtain the governing equations of the backward probability model for conservative tracers. The advantage of the adjoint approach over the heuristic approach in obtaining the governing equations is that adjoint theory provides a rigorous mathematical procedure for developing the equations for all aquifer geometries and for all transport processes. As additional processes are introduced to the problem, the adjoint equation must be rederived to ensure that the appropriate backward continuum model is obtained. Likewise, if random walk models are used, the correct backward equation must also be rederived if new processes are included.

We used adjoint theory to obtain backward location and travel time probabilities for the cases of spatially-varying porosity, transient flow and temporally-varying porosity, and distributed internal sources of solute and water. In each case, the backward governing equation was obtained directly from adjoint theory, indicating that the adjoint method is sufficiently robust to handle general spatially- and temporally-varying aquifer states and properties and distributed internal sources of solute and water. We illustrated each of these generalizations using simple one-dimensional vertically-integrated models. The approach is easily extended to multiple dimensions with irregular geometries using the approach of [17].

For non-constant porosity, we obtained more general forms of the adjoint equation and of the relationships between probabilities and adjoint states. The constant-porosity results of Neupauer and Wilson [16,17] are special cases of these. We found that spatially-varying porosity produces a non-uniform velocity field that affects both the movement and the spread of the probability distributions. We would observe similar results for variable aquifer thickness in vertically-integrated models or variable moisture content in unsaturated flow. With transient flow, the velocity field and porosity are spatially- and temporally-variable. The flow field in the backward model is reversed in both space and time relative to the flow field for the forward model.

A non-uniform velocity field can also result from areally-distributed natural recharge or other distributed internal sources or sinks of water. With internal sources of water, two interpretations of backward probability are possible. The first interpretation is used if contamination is known to enter through an internal source, such as natural recharge in a vertically integrated aquifer model. In the adjoint (backward) equation, the natural recharge term is equivalent to a first-order decay term; therefore, backward probabilities decay in time. For location probability at a fixed backward time, $\hat{\tau}$, this decay quantifies the probability that the observed contaminant parcel entered the aquifer through natural recharge during the time interval $0 < \tau < \hat{\tau}$, and was not in the system at $\hat{\tau}$. For travel time probability at a fixed point, $\hat{x}$, the decay indicates a non-zero probability that the contaminant parcel entered the system downgradient of $\hat{x}$, and therefore it was never at $\hat{x}$. The second interpretation is used if it is known that the source of contamination has no causal or correlative link to recharge. In this case, the backward probability can be calculated without decay by eliminating the recharge term from the adjoint equation. The choice of interpretation is based on the judgment of the modeler with knowledge of the physical system.

The backward probability model is an efficient method for obtaining information about the prior position of contamination that is observed in an aquifer. For each observation, only one simulation is needed to obtain information about all possible sources. In addition, the reversal of information can lead to new insight into the physical situation. For example, the two interpretations that are possible when internal sources of water are present show that the backward model can provide new kinds of information that can improve the understanding of the physical system.

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Appendix A. Adjoint derivation

To eliminate the state sensitivity, ψ, from (11), we first obtain a governing equation for ψ by differentiating each term of (6) with respect to the parameter $\alpha = M_0$, resulting in

$$-\frac{\partial}{\partial t}(\theta\psi)+\frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi}{\partial x}\right)-\frac{\partial}{\partial x}(\theta v\psi)-q_{\mathrm{O}}\psi=0, \quad \text{(A.1)}$$

$$\psi(x,0)=\frac{\partial C_{\mathrm{i}}(x)}{\partial M_0},$$

$$\frac{\partial\psi}{\partial x}=0 \quad \text{at } x=x_1,$$

$$\psi=0 \quad \text{at } x=x_2,$$

where we assume that g_1 and g_2 are independent of M_0 (although this assumption is not necessary), and by definition, the derivatives of D, v, θ, q_{I}, C_{I}, and q_{O} with respect to M_0 all vanish. Note that the water source term vanishes from this equation.

Next we take the inner product of each term of (A.1) with an arbitrary function, ψ^*, the adjoint state. The inner product in the space of continuous, square-integrable, real functions is $\langle f,g\rangle=\int_0^T\int_{x_1}^{x_2} f\,g\,\mathrm{d}x\,\mathrm{d}t$, where the time domain is $0\leqslant t\leqslant T$. Taking the inner product of each term in (A.1) with ψ^* produces

$$\int\int_{x,t}\left[-\psi^*\frac{\partial}{\partial t}(\theta\psi)+\psi^*\frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi}{\partial x}\right)-\psi^*\frac{\partial}{\partial x}(\theta v\psi)-\psi^* q_{\mathrm{O}}\psi\right]\mathrm{d}x\,\mathrm{d}t=0. \quad \text{(A.2)}$$

Using integration by parts on each derivative term, we can rewrite the inner product as

$$\int\int_{x,t}\left[\theta\psi\frac{\partial\psi^*}{\partial t}+\psi\frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi^*}{\partial x}\right)+\theta v\psi\frac{\partial\psi^*}{\partial x}-\psi^* q_{\mathrm{O}}\psi\right]\mathrm{d}x\,\mathrm{d}t-\int_x(\psi^*\theta\psi)\Big|_{t=0}^{t=T}\mathrm{d}x+\int_t\left[\psi^*\theta D\frac{\partial\psi}{\partial x}-\psi\theta D\frac{\partial\psi^*}{\partial x}-\psi^*\theta v\psi\right]\Bigg|_{x=x_1}^{x=x_2}\mathrm{d}t=0. \quad \text{(A.3)}$$

Since the left-hand side of this equation evaluates to zero, it can be added to the right-hand side of (11) without changing the equality. With this addition, after evaluation of the integrands of the single integrals at the boundaries, (11) becomes

$$\frac{\mathrm{d}P}{\mathrm{d}M_0}=\int\int_{x,t}\psi\left[\frac{\partial h}{\partial C}+\theta\frac{\partial\psi^*}{\partial t}+\frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi^*}{\partial x}\right)+\theta v\frac{\partial\psi^*}{\partial x}-q_{\mathrm{O}}\psi^*\right]\mathrm{d}x\,\mathrm{d}t-\int_x\left[(\psi^*\theta\psi)\big|_{t=T}-(\theta\psi^*)\big|_{t=0}\frac{\partial C_{\mathrm{i}}}{\partial M_0}\right]\mathrm{d}x+\int_t\left[\psi^*\theta D\frac{\partial\psi}{\partial x}\bigg|_{x=x_2}+\psi\theta\left(D\frac{\partial\psi^*}{\partial x}+v\psi^*\right)\bigg|_{x=x_1}\right]\mathrm{d}t. \quad \text{(A.4)}$$

To eliminate ψ from the equation, we define the adjoint state, ψ^*, such that the terms containing ψ vanish. The double integral can be eliminated if the terms inside the square brackets sum to zero; this produces the adjoint equation

$$-\theta\frac{\partial\psi^*}{\partial t}=\frac{\partial}{\partial x}\left(\theta D\frac{\partial\psi^*}{\partial x}\right)+\theta v\frac{\partial\psi^*}{\partial x}-q_{\mathrm{O}}\psi^*+\frac{\partial h}{\partial C}, \quad \text{(A.5)}$$

with ψ^* as the adjoint state. Using the continuity equation for an incompressible fluid,

$$\frac{\partial\theta}{\partial t}+\frac{\partial}{\partial x}(\theta v)=q_{\mathrm{I}}-q_{\mathrm{O}}, \quad \text{(A.6)}$$

the adjoint equation can be rewritten as (13).

All but one of the remaining terms in (A.4) vanish if $D(\partial\psi^*/\partial x)+v\psi^*=0$ at $x=x_1$, $\psi^*=0$ at $x=x_2$, and $\psi^*=0$ at $t=T$. These are the final and boundary conditions for the adjoint state, ψ^*, in (13). With this definition of the adjoint state, the marginal sensitivity in (A.4) is simplified to the expression in (12).

Appendix B. Backward probability simulations using MT3DMS with transient flow

For transient flow of an incompressible fluid, temporal variability in hydraulic head causes temporal variability in porosity, given by

$$S_{\mathrm{s}}\frac{\partial h}{\partial t}=\frac{\partial\theta}{\partial t}, \quad \text{(B.1)}$$

where S_{s} is specific storage, h is head, and θ is porosity. Although most flow codes account for small changes in water storage through S_{s}, few flow and transport models account for the actual change in aquifer thickness or change in porosity. In general, this inconsistency produces small mass balance errors in the transport model that are almost always ignored. To verify that the backward probability model is correctly developed, we require that probability (equivalent to mass in a forward contaminant transport model) be preserved. Although this inconsistency in the general treatment of storage effects prohibits a complete verification of the backward probability model, we made a small change in our use of MT3DMS to ensure mass conservation.

In MT3DMS [26], the accumulation term in (6) is separated into

$$\frac{\partial(\theta C)}{\partial t}=\theta\frac{\partial C}{\partial t}+C\frac{\partial\theta}{\partial t}=\theta\frac{\partial C}{\partial t}+Cq_{\mathrm{s}}', \quad \text{(B.2)}$$

where $q_{\mathrm{s}}'=\partial\theta/\partial t$ is the "flow rate" (per unit volume) of water into or out of storage. The values of q_{s}' are calculated in the flow simulation (MODFLOW-96) as $S_{\mathrm{s}}(\partial h/\partial t)$, and are entered into MT3DMS via the MODFLOW/MT3D link file. The term containing q_{s}' implicitly accounts for the time-variability of the pore volume; however, porosity in the $\theta(\partial C/\partial t)$ term is not updated from its original value. This leads to an inconsistent treatment of the time-variability of porosity,

producing usually negligible mass balance errors. To preserve mass and probability in our examples, we set $q'_s = 0$ for all forward and backward transient transport simulations. The specific discharge, q, is still transient, but the time-variability of porosity is eliminated.

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