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Adjoint method for obtaining backward-in-time location and travel time probabilities of a conservative groundwater contaminant

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Abstract. Backward location and travel time probabilities can be used to determine the prior location of contamination in an aquifer. For a contaminant particle that was detected in an aquifer, the backward location probability is the probability of where the particle was located at some prior time. Backward travel time probability is the probability of when the particle was located at some position upgradient of the detection. These probabilities can be used to improve characterization of known sources of groundwater contamination, to identify previously unknown contamination sources, and to delineate capture zones. For simple model domains, backward probabilities can be obtained heuristically from a forward model of contaminant transport. For multidimensional problems and complex domain geometries, the heuristic approach is difficult to implement and verify. The adjoint method provides a formal approach for obtaining backward probabilities for all model domains and geometries. We formally show that the backward model probabilities are adjoint states of resident concentration. We provide a methodology for obtaining the governing equations and boundary and final conditions for these probabilities. The approach is illustrated using a one-dimensional, semi-infinite domain that mimics flow to a production well, and these results are compared to equivalent probabilities derived heuristically.

1. Introduction

Transport of a conservative solute in groundwater is usually described by the advection-dispersion equation (ADE). Solutions of the ADE express solute concentration as a function of location and time, for all times after the initial release of the solute. This form of the ADE is a forward-in-time model because we track the solute as it moves forward in time.

Concentration can be expressed in two different forms: resident concentration and flux concentration [*Kreft and Zuber*, 1978; *Parker and van Genuchten*, 1984]. Resident concentration is a measure of the mass of solute per unit volume of water, or a volume-averaged concentration. Flux concentration is a measure of the solute mass flux per unit water flux or a flux-averaged concentration. The forward-in-time (forward) ADE can be solved for either resident or flux concentration.

The forward ADE can also be used to solve for location probability and travel time probability. If we consider an individual solute parcel that was released from the contaminant source, then the location probability of that parcel is the probability that it is located at a given position in space at some later time [*Dagan*, 1989; *Jury and Roth*, 1990; *Chin and Chittaluru*, 1994]. Location probability is related to resident concentration [*Jury and Roth*, 1990]. Resident concentration measures the mass of solute at a given location in space at a snapshot in time. If the resident concentration measurements are normalized by the total mass of solute in the system, the resulting distribution is the percentage of the total mass that is at a given location in space. Suppose we are interested in the present location of one parcel of mass that was input at the source. The parcel is more likely to be found at a location that has a high solute concentration (or, equivalently, a high normalized solute concentration) than a location that has a low solute concentration. Thus, at any point in time, the normalized concentration distribution is equivalent to a probability density function for the location of the parcel (i.e., location probability). Note that for a unit source, the resulting resident concentration is equal to the location probability.

If we again consider an individual solute parcel that was released from the contaminant source, then the travel time probability of that parcel is the probability that it will arrive at a fixed location after a given amount of time has elapsed [*Jury*, 1982; *Chin and Chittaluru*, 1994]. Travel time probability is related to flux concentration [*Jury*, 1982]. Flux concentration measures the mass of solute passing through a fixed location in space over a finite time interval. Mass flux can be expressed as the product of flux concentration and groundwater velocity. In a one-dimensional system, the entire mass of a conservative solute must eventually pass through every point downstream of the source. For a given location downstream of the source, if the mass flux is normalized by the total mass released from the source, the resulting distribution shows the percentage of the total mass that passes the given location in any finite time interval. Suppose we are interested in determining when one parcel of mass that was input at the source will reach a given location. This parcel is more likely to reach the given location when the flux concentration (or normalized mass flux) is high rather than when it is low. Therefore, for a solute parcel traveling from the source to any given location, the normalized mass flux distribution is equivalent to the travel time probability density function. Since these forward-in-time probabilities are equivalent to normalized concentrations, the forward ADE can also be solved for location and travel time probabilities.

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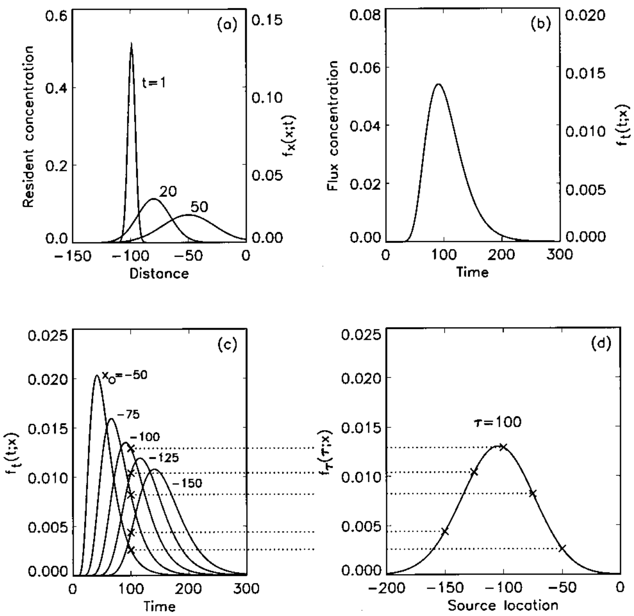

Figure 1. Plots of location and travel time probability, similar to plots presented by *Wilson and Liu* [1995]. (a) Plot of resident concentration and forward location probability for a contamination source at $x_o = -100$ for three times. (b) Plot of flux concentration at the pumping well (at $x = 0$) and forward travel time probability to the pumping well for a contamination source at $x_o = -100$. (c) Plot of forward travel time probability to the pumping well for five different contamination source locations. The cross denotes the probability that $\tau = 100$ for each source location. (d) Plot of backward travel time probability from the pumping well, showing the probability that $\tau = 100$ for all possible source locations. The cross denotes $f_\tau(\tau; x)$ for the source locations in Figure 1c.

In this discussion, the probabilities have been defined from the point of view of the contaminant source; i.e., they are probabilities for a solute parcel that originated at the contaminant source. Suppose we detect contaminant in the groundwater, but its prior location and source is unknown. We can use a backward-in-time version of the ADE to solve for the prior location probability of the detected solute parcel and also for the travel time probability of the detected solute parcel from some upgradient location to the detection location [*Wilson and Liu*, 1995]. These probabilities can be used to improve characterization of known sources of groundwater contamination, to identify previously unknown contamination sources, and to delineate capture zones. A related problem that has received much attention recently is a deterministic problem in which the release history of a contamination source is reconstructed from the present position of the contamination plume [e.g., *Skaggs and Kabala*, 1994]. In the source history reconstruction problem, the source location is assumed to be known, while the problem presented here can be used to obtain information about the locations of unknown sources.

In the forward model, we are interested in where the solute parcel is going, so the flow of information is away from the source, i.e., in the direction of the groundwater velocity. In the backward model, we are interested in where the solute parcel has been, so the flow of information is away from the detection and back toward the (possibly unknown) source, i.e., in the opposite direction as the groundwater velocity. Given a forward model, the equivalent backward probability model can be obtained heuristically by reversing the sign on the advection term to account for the reversed flow of information and by modifying the source term, boundary conditions, and initial condition to include information about the detected solute parcel. Although the dispersion coefficient is a function of velocity, no sign reversal is performed on the dispersion term. Dispersion is proportional to the magnitude of velocity; therefore reversing the direction of velocity does not affect the sign on the dispersion coefficient. The resulting backward-in-time location and travel time probabilities can provide information about the prior location of contamination before it was detected in the aquifer. *Wilson and Liu* [1995], who first derived expressions for backward-in-time location and travel time probabilities, used this approach. Although they heuristically obtained accurate expressions for these probabilities (as demonstrated in our Figure 1), no formal justification was given for the governing equation or boundary conditions. Furthermore, the heuristic approach is difficult to implement and verify for multidimensional problems and complex domain geometries.

Bagtzoglou et al. [1992] also used backward location probabilities to identify sources of contamination. They obtained probability maps using a random walk method by reversing the flow field and leaving the dispersion process unchanged. *Uffink* [1989] used a similar random walk approach to delineate capture zones around pumping wells.

We propose the adjoint method as a formal approach for obtaining backward probabilities for all model domains. The adjoint method has been used in a variety of applications in groundwater modeling including sensitivity analysis [*Sykes et al.*, 1985; *Wilson and Metcalfe*, 1985], parameter estimation [*Neuman*, 1980; *Sun and Yeh*, 1985; *Townley and Wilson*, 1985; *Lu et al.*, 1988; *Sun and Yeh*, 1990; *Yeh and Sun*, 1990], optimal design [*Ahlfeld et al.*, 1988], and others [see *Sun*, 1994]. Central to each of these methods is a linear functional known as a performance measure or an objective function. The performance measure is problem specific and depends on the goal of the study. For example, in their sensitivity analysis, *Sykes et al.* [1985] defined one of their performance measures to be piezometric head at a point in an aquifer and then used the adjoint method to determine sensitivity of this performance measure to the model boundary conditions. *Sun and Yeh* [1985], in a parameter estimation problem, defined their performance measure to be the sum of the squared differences between measured and modeled piezometric head values. They used the adjoint method to determine the sensitivity of this performance measure to the aquifer transmissivity values and then used these results to find the optimal values.

With the adjoint method, the forward governing equation (forward operator), with concentration as the dependent variable, is replaced by the adjoint equation (adjoint operator), with the adjoint state as the dependent variable. The adjoint state is a function that describes the marginal change in the performance measure due to a unit injection of mass at any point in the system, as we will illustrate later. A different adjoint state can be defined for each performance measure. The adjoint equation models the same physical processes as the forward equation; however, the flow of information is reversed (i.e., the adjoint state is propagated backward in time). A Green's function is an example of an adjoint state.

In this paper, we show that backward-in-time location and travel time probabilities are adjoint states of forward-in-time resident concentration. We provide a methodology for obtaining the governing equations and boundary and initial conditions for these probabilities and show that they are equivalent to those derived by *Wilson and Liu* [1995]. The illustration is shown for a one-dimensional, semi-infinite domain. Although one-dimensional flow and transport is highly idealized, it is equivalent to the domain used by *Wilson and Liu* [1995], and after the methodology has been developed and verified for a one-dimensional system, we can easily extend it to multiple dimensions. The one-dimensional, semi-infinite domain represents flow and transport to a production well. The production well is the driving force for flow and acts as a sink of water and solute. The adjoint approach can be applied to other domains, such as an infinite domain with an interior monitoring well or weak production well as a detection mechanism. In these infinite domain scenarios, the driving force for flow would be a prescribed velocity field.

In the next section, we present forward models for resident and flux concentration, and we show the relationship between these models and the corresponding forward probabilities. The backward models for location and travel time probabilities are developed heuristically from the forward probability models, following the work of *Wilson and Liu* [1995] and *Liu* [1995]. In the subsequent section, a general form of the adjoint equation is derived. It is then applied to the semi-infinite domain to derive two different adjoint states, one representing location probability and the other representing travel time probability. We show that these probabilities are equivalent to the backward model probabilities that were developed heuristically by *Wilson and Liu* [1995].

2. One-Dimensional Contaminant Transport

In this section, solutions to the advection-dispersion equation are described for resident and flux concentration in a one-dimensional system for an instantaneous point source of contaminant. Using these equations, expressions are developed for location and travel time probabilities for both forward and backward models.

Wilson and Liu [1995] developed expressions for backward-in-time location and travel time probabilities in a one-dimensional, semi-infinite domain. In that work, the domain extended from $0 \leq x < \infty$, with a pumping well (detection mechanism) at $x = 0$ and an instantaneous point source of contaminant at $x_o > 0$. Thus the velocity was in the direction of $-x$. The equations presented here are taken from *Wilson and Liu* [1995]; however, in this work, the domain extends from $-\infty < x \leq 0$, and velocity is moving in the $+x$ direction. The pumping well is still located at $x = 0$, and the instantaneous point source of contaminant is at $x_o < 0$.

2.1. Forward Model

Contaminant transport in a one-dimensional, semi-infinite domain is described by the following form of the advection-dispersion equation:

$$\frac{\partial C^r}{\partial t} = \frac{\partial}{\partial x}\left(D\,\frac{\partial C^r}{\partial x}\right) - \frac{\partial(vC^r)}{\partial x} \tag{1}$$

$$C^r \rightarrow 0 \qquad x \rightarrow -\infty$$

$$\frac{\partial C^r}{\partial x} = 0 \qquad x = 0$$

$$C^r(x,\,0) = \frac{M}{A\theta}\,\delta(x - x_o),$$

where C^r is resident concentration, D is the dispersion coefficient, v is groundwater velocity, x is the spatial dimension, t is time, M is total source mass, A is cross-sectional area, θ is porosity, $\delta(x)$ is the Dirac delta function, and x_o is the source location ($x_o < 0$). The boundary at $x = 0$ represents a pumping well. The boundary condition specifies that the concentration inside the well bore is equal to the concentration of the fluid in the porous media immediately adjacent to the well bore. Although this might not be exact, it is mathematically convenient and an accepted boundary condition at a pumping well [e.g., *Chen and Woodside*, 1988].

Wilson and Liu [1995] derived the solution to this problem for constant v and D:

$$C^r(x,\,t) = \frac{1}{\sqrt{4\pi Dt}}\,\frac{M}{A\theta}\exp\left\{-\frac{(x - x_o - vt)^2}{4Dt}\right\} \cdot \left[1 + \exp\left\{\frac{-x_o x}{Dt}\right\}\right] - \frac{v}{2D}\,\frac{M}{A\theta}\exp\left\{\frac{-vx_o}{D}\right\}\,\mathrm{erfc}\left[\frac{-x - x_o + vt}{\sqrt{4Dt}}\right]. \tag{2}$$

By normalizing resident concentration by the total mass in the system, we obtain the following expression for location probability, $f_x(x;\,t)$:

$$f_x(x;\,t) = \frac{C^r(x,\,t)}{\displaystyle\int_{-\infty}^{0} C^r(x,\,t)\,dx}. \tag{3}$$

The integral in the denominator evaluates to $M/A\theta$, which can be obtained by integrating (2). Alternatively, we see from (1) that there are no internal sources or sinks of contamination, and the contaminant is nonreactive; therefore mass is conserved. In other words, the total mass in the system at any time is equal to the total mass in the system at the initial time. Since $C^r(x,\,0) = (M/A\theta)\delta(x - x_o)$, where $-\infty < x_o \leq 0$, the total mass in the system at any time is $\int_{-\infty}^{0} C^r(x,\,0)\,dx = M/A\theta$; so location probability in (3) is equal to $C^r/(M/A\theta)$, and

$$f_x(x;\,t) = \frac{1}{\sqrt{4\pi Dt}}\exp\left\{-\frac{(x - x_o - vt)^2}{4Dt}\right\} \cdot \left[1 + \exp\left\{\frac{-x_o x}{Dt}\right\}\right] - \frac{v}{2D}\exp\left\{\frac{-vx_o}{D}\right\}\,\mathrm{erfc}\left[\frac{-x - x_o + vt}{\sqrt{4Dt}}\right]. \tag{4}$$

This equation could also be obtained by solving (1), replacing C^r with f_x, and using the initial condition $f_x(x;\,0) = \delta(x - x_o) = C^r(x,\,0)/(M/A\theta)$.

Figure 1a shows plots of resident concentration and location probability as a function of position for three different times, $t = 1$, 20, and 50 (dimensionless units). The source location is at $x_o = -100$. Other parameter values are $v = 1.0$, $D = 5.0$, $M = 1.0$, $A = 1.0$, and $\theta = 0.25$. These parameter values were used for all plots in Figure 1. A high value of the dispersion coefficient was used to illustrate the effects of dispersion. The left-hand axis represents resident concentration, and the right-hand axis represent location probability (resident concentration normalized by $M/A\theta = 4.0$).

Flux concentration is related to resident concentration as follows [*Parker and van Genuchten*, 1984]:

$$C^f = C^r - \frac{D}{v}\frac{\partial C^r}{\partial x}, \tag{5}$$

where C^f is flux concentration. For the semi-infinite domain described above, the gradient of C^r at $x = 0$ is zero; therefore, at the pumping well ($x = 0$), flux concentration is equal to resident concentration [*Wilson and Liu*, 1995]

$$C^f(0, t) = \frac{1}{\sqrt{\pi D t}}\frac{M}{A\theta}\exp\left\{-\frac{(x_o + vt)^2}{4Dt}\right\} - \frac{v}{2D}\frac{M}{A\theta}\exp\left\{\frac{-vx_o}{D}\right\}\operatorname{erfc}\left[\frac{-x_o + vt}{\sqrt{4Dt}}\right]. \tag{6}$$

Normalizing flux concentration by the total mass in the system results in the following expression for travel time probability, $f_t(t; x)$, for transport from the source to the well:

$$f_t(t; x) = \frac{vC^f(x, t)}{\int_0^\infty vC^f(x, t)\, dt}. \tag{7}$$

Integrating the first term in (6) over the time domain yields $(2/v)(M/A\theta)$ [*Abramowitz and Stegun*, 1972, equation 29.3.84] and integrating the second term in (6) yields $-(1/v)(M/A\theta)$ [*Abramowitz and Stegun*, 1972, equation 7.4.21]; thus the integral in the denominator of (7) evaluates to $M/A\theta$. The travel time probability is equal to $vC^f/(M/A\theta)$, and

$$f_t(t; 0) = \frac{v}{\sqrt{\pi D t}}\exp\left\{-\frac{(x_o + vt)^2}{4Dt}\right\} - \frac{v^2}{2D}\exp\left\{\frac{-vx_o}{D}\right\}\operatorname{erfc}\left[\frac{-x_o + vt}{\sqrt{4Dt}}\right]. \tag{8}$$

Figure 1b shows a plot of flux concentration and travel time probability at the pumping well at $x = 0$, as a function of time, for contamination from a source at location $x_o = -100$. The left-hand axis represents flux concentration, and the right-hand axis represents travel time probability (flux concentration normalized by $M/A\theta = 4.0$).

2.2. Backward Model

For constant D and v, *Wilson and Liu* [1995] showed that the backward model for location probability can be obtained by solving the following equation:

$$\frac{\partial f_x}{\partial \tau} = D\frac{\partial^2 f_x}{\partial x^2} + v\frac{\partial f_x}{\partial x} \tag{9}$$

$$f_x \to 0 \qquad x \to -\infty$$

$$vf_x + D\frac{\partial f_x}{\partial x} = 0 \qquad x = 0$$

$$f_x(x; 0) = \delta(x - x_d),$$

where τ is backward time ($\tau = t_d - t$, where t_d is the detection time) and x_d is the detection location, which here is the location of the pumping well at $x_d = 0$. The solution to this equation is the backward-in-time location probability for a detection at $x_d = 0$ and is given by [*Wilson and Liu*, 1995]

$$f_x(x; \tau) = \frac{1}{\sqrt{\pi D\tau}}\exp\left\{-\frac{(x + v\tau)^2}{4D\tau}\right\} - \frac{v}{2D}\exp\left\{\frac{-vx}{D}\right\}\operatorname{erfc}\left[\frac{-x + v\tau}{\sqrt{4D\tau}}\right]. \tag{10}$$

For a contaminant parcel removed at the pumping well ($x_d = 0$), this equation describes the probability density function of the location of the contaminant parcel at time τ before it was observed at the pumping well. *Wilson and Liu* [1995] also showed that the backward model for travel time probability can be obtained by solving

$$\frac{\partial f_\tau}{\partial \tau} = D\frac{\partial^2 f_\tau}{\partial x^2} + v\frac{\partial f_\tau}{\partial x} \tag{11}$$

$$f_\tau \to 0 \qquad x \to -\infty$$

$$vf_\tau + D\frac{\partial f_\tau}{\partial x} = v\delta(\tau) \qquad x = 0$$

$$f_\tau(x; 0) = 0$$

where D and v are constant and the pumping well is assumed to be at $x_d = 0$. The solution to this equation is the backward-in-time travel time probability from location x to a detection at $x_d = 0$ [*Wilson and Liu*, 1995].

$$f_\tau(\tau; x) = \frac{v}{\sqrt{\pi D\tau}}\exp\left\{-\frac{(x + v\tau)^2}{4D\tau}\right\} - \frac{v^2}{2D}\exp\left\{\frac{-vx}{D}\right\}\operatorname{erfc}\left[\frac{-x + v\tau}{\sqrt{4D\tau}}\right]. \tag{12}$$

For a contaminant parcel removed at the pumping well, this equation describes the probability density function of the travel time from some upgradient location x to the pumping well at $x_d = 0$.

Figures 1c and 1d provide an illustration of the relationship between forward and backward travel-time-probability. Figure 1c shows the forward travel time probability from many source locations to the pumping well at $x = 0$. For a travel time of $t = 100$, the travel time probability is denoted with a cross. For a backward time of $\tau = 100$, Figure 1d shows the backward travel time probability from the pumping well for all possible source locations. The source locations corresponding to those shown in Figure 1c are indicated with a cross. These plots show that the backward probabilities can be obtained from the multiple forward probabilities; however, more simulations are needed (one for each possible source location), and less information is obtained (probability is only obtained for a finite number of source locations).

3. Derivation of Adjoint Equations

A common application of the adjoint method [*Marchuk et al.*, 1996] in groundwater hydrology is sensitivity analysis [e.g., *Sykes et al.*, 1985; *Wilson and Metcalfe*, 1985; *Sun and Yeh*, 1990], often in the context of an inverse problem. Sensitivity analysis is used to determine the sensitivity of the state of the system (model output) to changes in parameter values (model input). The direct method of performing a sensitivity analysis is to vary the input parameter slightly, rerun the model, and determine the effect on the model output. This method requires one simulation for each parameter. The adjoint method offers a more efficient approach in which the adjoint equation is solved once, and then the result is used to directly compute the sensitivity of the state of the system to all parameters.

Although we are not interested in performing a sensitivity analysis, we follow the sensitivity analysis approach to obtain the adjoint equation. In this section, the general adjoint equation is derived for the advection-dispersion equation. Then, the derivation is applied to location and travel time probabilities in a semi-infinite domain. By first developing the general adjoint equation, we can adapt the results to address other model domains, such as the infinite domain problem, which could simulate flow and transport through an observation well.

3.1. General Adjoint Equation

The adjoint of the advection-dispersion equation (ADE) is developed here following the sensitivity analysis approach of *Sykes et al.* [1985] (see also *Sun and Yeh* [1990] and *Sun* [1994]). In sensitivity analysis, a performance measure is defined that quantifies the state of the system. The goal is to determine the marginal sensitivity of this performance measure to small changes in parameter values. In this section, the adjoint equation is first derived for the advection-dispersion equation using a general performance measure.

The performance measure P that quantifies the state of the system can be expressed as

$$P = \int\int_{x,t} h(\alpha, C)\, dx\, dt, \tag{13}$$

where $h(\alpha, C)$ is a functional of the state of the system, α is a vector of system parameters (e.g., $\alpha = [v, D, M, \theta]$), C is concentration, and the integration is over the entire space-time domain. The marginal sensitivity of this performance measure with respect to one parameter, α_k, is obtained by differentiating (13) with respect to α_k:

$$\frac{dP}{d\alpha_k} = \int\int_{x,t} \left[\frac{\partial h(\alpha, C)}{\partial \alpha_k} + \frac{\partial h(\alpha, C)}{\partial C}\, \psi \right] dx\, dt, \tag{14}$$

where $dP/d\alpha_k$ is the marginal sensitivity and ψ is the state sensitivity, $\psi = \partial C/\partial \alpha_k$, where ψ is a measure of the change in system state, C, due to a small change in one of the parameters, α_k, while holding constant x, t, and the other parameters in α. Since the state sensitivity is unknown, adjoint theory can be used to eliminate it from the previous equation. This is done by first differentiating the advection-dispersion equation (including initial and boundary conditions) with respect to the parameter α_k, to obtain a form of the ADE in terms of the state sensitivity, ψ.

One-dimensional contaminant transport can be described by the following general form of the advection-dispersion equation:

$$-\frac{\partial C}{\partial t} + \frac{\partial}{\partial x}\left(D \frac{\partial C}{\partial x} \right) - \frac{\partial (vC)}{\partial x} + Q(x, t) = 0, \tag{15}$$

where $Q(x, t)$ describes the contaminant source. The general initial and boundary conditions for this equation are

$$a_1 C + b_1 \frac{\partial C}{\partial x} = g_1(t) \qquad x = x_1$$

$$a_2 C + b_2 \frac{\partial C}{\partial x} = g_2(t) \qquad x = x_2$$

$$C(x, 0) = g_3(x),$$

where x_1 and x_2 are the boundary locations, $x_2 > x_1$, while a_1, a_2, b_1, and b_2 are known constants, and $g_1(t)$, $g_2(t)$, and $g_3(x)$ are known functions.

Differentiating (15) and its boundary and initial conditions with respect to α_k gives

$$-\frac{\partial \psi}{\partial t} + \frac{\partial}{\partial x}\left(D \frac{\partial \psi}{\partial x} \right) + \frac{\partial}{\partial x}\left(\frac{\partial D}{\partial \alpha_k} \frac{\partial C}{\partial x} \right) - \frac{\partial (v\psi)}{\partial x} - \frac{\partial}{\partial x}\left(C \frac{\partial v}{\partial \alpha_k} \right) + \frac{\partial Q(x, t)}{\partial \alpha_k} = 0 \tag{16}$$

$$a_1 \psi + b_1 \frac{\partial \psi}{\partial x} = \frac{\partial g_1(t)}{\partial \alpha_k} \qquad x = x_1$$

$$a_2 \psi + b_2 \frac{\partial \psi}{\partial x} = \frac{\partial g_2(t)}{\partial \alpha_k} \qquad x = x_2$$

$$\psi(x, 0) = \frac{\partial g_3(x)}{\partial \alpha_k},$$

where we assumed that the coefficients a_1, a_2, b_1, and b_2 are independent of α_k, appropriate for the problems presented in this paper.

The next step is to obtain a similar form of the ADE in terms of the adjoint state, ψ^*, which, at this stage, is just an arbitrary function. First, we define the inner product of two functions, ψ^* and ξ, to be $\int\int_{x,t} \psi^* \bar{\xi}\, dx\, dt$, where the overbar denotes complex conjugate. All functions used in this paper are real; therefore $\bar{\xi} = \xi$. Taking the inner product of the adjoint state, ψ^*, and each term on both sides of (16) gives

$$\int_0^T \int_{x_1}^{x_2} \left[-\psi^* \frac{\partial \psi}{\partial t} + \psi^* \frac{\partial}{\partial x}\left(D \frac{\partial \psi}{\partial x} \right) - \psi^* \frac{\partial (v\psi)}{\partial x} + \psi^* \frac{\partial Q(x, t)}{\partial \alpha_k} + \psi^* \frac{\partial}{\partial x}\left(\frac{\partial D}{\partial \alpha_k} \frac{\partial C}{\partial x} \right) - \psi^* \frac{\partial}{\partial x}\left(C \frac{\partial v}{\partial \alpha_k} \right) \right] dx\, dt = 0. \tag{17}$$

Integration is carried out over the entire domain: $x_1 \le x \le x_2$, and $0 \le t \le T$ (we will later show that the final time T is equivalent to the detection time). This equation can be manipulated, term by term, to obtain a similar form of the ADE with ψ^* as the state variable, and with additional divergence terms. Take the first term

$$\int_0^T \int_{x_1}^{x_2} -\psi^* \frac{\partial \psi}{\partial t}\, dx\, dt = \int_0^T \int_{x_1}^{x_2} \left[-\frac{\partial}{\partial t}(\psi^* \psi) + \psi \frac{\partial \psi^*}{\partial t} \right] dx\, dt. \tag{18}$$

For the second term,

$$\int_0^T \int_{x_1}^{x_2} \psi^* \frac{\partial}{\partial x}\left(D \frac{\partial \psi}{\partial x} \right) dx\, dt = \int_0^T \int_{x_1}^{x_2} \left[\frac{\partial}{\partial x}\left(D\psi^* \frac{\partial \psi}{\partial x} \right) - D \frac{\partial \psi^*}{\partial x} \frac{\partial \psi}{\partial x} \right] dx\, dt$$

$$= \int_0^T \int_{x_1}^{x_2} \left[\frac{\partial}{\partial x}\left(D\psi^* \frac{\partial \psi}{\partial x} \right) - \frac{\partial}{\partial x}\left(D\psi \frac{\partial \psi^*}{\partial x} \right) + \psi \frac{\partial}{\partial x}\left(D \frac{\partial \psi^*}{\partial x} \right) \right] dx\, dt. \tag{19}$$

The third term becomes

$$\int_0^T \int_{x_1}^{x_2} -\psi^* \frac{\partial(v\psi)}{\partial x}\, dx\, dt$$

$$= \int_0^T \int_{x_1}^{x_2} \left[-\frac{\partial}{\partial x}(v\psi^*\psi) + \psi v \frac{\partial \psi^*}{\partial x} \right] dx\, dt. \tag{20}$$

No manipulation is done to the remaining terms in (17).

Substituting (18), (19), and (20) into (17) and rearranging the terms gives

$$\int_0^T \int_{x_1}^{x_2} \left\{ \psi \left[\frac{\partial \psi^*}{\partial t} + \frac{\partial}{\partial x}\left(D \frac{\partial \psi^*}{\partial x} \right) + v \frac{\partial \psi^*}{\partial x} \right] + \psi^* \frac{\partial Q}{\partial \alpha_k} + \psi^* \frac{\partial}{\partial x}\left(\frac{\partial D}{\partial \alpha_k} \frac{\partial C}{\partial x} \right) - \psi^* \frac{\partial}{\partial x}\left(C \frac{\partial v}{\partial \alpha_k} \right) + \frac{\partial}{\partial x} \cdot \left[D\psi^* \frac{\partial \psi}{\partial x} - D\psi \frac{\partial \psi^*}{\partial x} - v\psi^*\psi \right] - \frac{\partial}{\partial t}(\psi^*\psi) \right\} dx\, dt = 0. \tag{21}$$

Since the left-hand side of this equation is equal to zero, it can be added to the right-hand side of the marginal sensitivity equation (14), yielding

$$\frac{dP}{d\alpha_k} = \int_0^T \int_{x_1}^{x_2} \left\{ \frac{\partial h(\alpha, C)}{\partial \alpha_k} + \psi \left[\frac{\partial h}{\partial C} + \frac{\partial \psi^*}{\partial t} + \frac{\partial}{\partial x}\left(D \frac{\partial \psi^*}{\partial x} \right) + v \frac{\partial \psi^*}{\partial x} \right] + \psi^* \frac{\partial Q}{\partial \alpha_k} + \psi^* \frac{\partial}{\partial x}\left(\frac{\partial D}{\partial \alpha_k} \frac{\partial C}{\partial x} \right) - \psi^* \frac{\partial}{\partial x}\left(C \frac{\partial v}{\partial \alpha_k} \right) + \frac{\partial}{\partial x} \cdot \left[D\psi^* \frac{\partial \psi}{\partial x} - D\psi \frac{\partial \psi^*}{\partial x} - v\psi^*\psi \right] - \frac{\partial}{\partial t}(\psi^*\psi) \right\} dx\, dt. \tag{22}$$

The last two terms in this equation are divergence terms, which, after integration, are evaluated at the boundary conditions. Thus these terms can be simplified as follows:

$$\int_0^T \int_{x_1}^{x_2} \frac{\partial}{\partial x} \left[D\psi^* \frac{\partial \psi}{\partial x} - D\psi \frac{\partial \psi^*}{\partial x} - v\psi^*\psi \right] dx\, dt = \int_0^T \left[D\psi^* \frac{\partial \psi}{\partial x} - D\psi \frac{\partial \psi^*}{\partial x} - v\psi^*\psi \right] \Bigg|_{x_1}^{x_2} dt \tag{23}$$

$$\int_0^T \int_{x_1}^{x_2} -\frac{\partial}{\partial t}(\psi^*\psi)\, dx\, dt = \int_{x_1}^{x_2} -(\psi^*\psi) \Bigg|_0^T dx \tag{24}$$

where the time domain extends from $t = 0$ to $t = T$.

Recall that the intent of this exercise is to eliminate the unknown state sensitivity, ψ, from the (14), or, equivalently, from (22). Recall also that the adjoint state, ψ^*, is still an arbitrary function. Thus the adjoint state, ψ^*, can be defined in such a way as to eliminate the state sensitivity, ψ, from (22). From these considerations, the governing equation for the adjoint state is

$$\frac{\partial h(\alpha, C)}{\partial C} + \frac{\partial \psi^*}{\partial t} + \frac{\partial}{\partial x}\left(D \frac{\partial \psi^*}{\partial x} \right) + v \frac{\partial \psi^*}{\partial x} = 0, \tag{25}$$

and the following statements must be satisfied by the boundary and initial conditions, respectively:

$$\left[D\psi^* \frac{\partial \psi}{\partial x} - D\psi \frac{\partial \psi^*}{\partial x} - v\psi^*\psi \right] \Bigg|_{x_1}^{x_2} = 0 \tag{26}$$

$$(\psi^*\psi)\big|_0^T = 0. \tag{27}$$

The boundary and initial conditions of the state sensitivity, ψ, have been defined in the governing equation of the forward model (16). By substituting these values for ψ into (26) and (27), we obtain the boundary and final conditions on ψ^*, in terms of known quantities (e.g., v and D). We have now defined an adjoint equation (25) and its boundary (26) and final (27) conditions, which can be solved to obtain the (no longer arbitrary) functional form of the adjoint state ψ^*. This function ψ^* is an adjoint state of the original state variable C.

Through this derivation, we see that there are many different adjoint states of C. For each definition of the performance measure, we have a different functional h and therefore different forms of the adjoint equation and the adjoint state. Two possible adjoint states are location probability and travel-time probability, which we derive in the next section. Also, in deriving the adjoint equation, we added zero (21) to the marginal sensitivity equation (14) to obtain an equivalent marginal sensitivity equation (22) from which we eliminated the state sensitivity ψ by defining an adjoint equation. We could have added a different form of zero (e.g., by multiplying (21) by a constant) to the marginal sensitivity equation. The result would be a different, but equally valid, adjoint equation.

Our intent was to obtain the adjoint of the ADE, which we have done in general form for one dimension. For completeness, we now discuss the relationship between adjoint states and sensitivity analysis. Each adjoint state represents a different measure of the system sensitivity. If we were performing a sensitivity analysis, we would use the appropriate adjoint state in the reduced form of (22) to obtain our solution to the marginal sensitivity, $dP/d\alpha_k$. Note that if we were performing a sensitivity analysis with respect to the source strength at one point in space-time, the adjoint state would be equivalent to the marginal sensitivity. In other words, let the contaminant source be an instantaneous point source at $x = x^*$ and $t = t^*$, so $Q(x, t) = Q^*\delta(x - x^*)\delta(t - t^*)$, where Q^* is the source strength. Then, α_k is the source strength, Q^*. Since f, D, and v are independent of Q^*, the terms in (22) containing their derivatives with respect to α_k are equal to zero. The terms in (22) that contain ψ are equal to zero by the definition of the adjoint state, ψ^*. Therefore the only nonzero term in (22) is $\psi^*\partial Q/\partial \alpha_k$. Since α_k is defined as Q^*, it can be seen that $\partial Q/\partial \alpha_k$ is equal to $\delta(x - x^*)\delta(t - t^*)$. Thus we have $\psi^*\partial Q/\partial \alpha_k = \psi^*\delta(x - x^*)\delta(t - t^*)$ and, from (22), $dP/d\alpha_k = \psi^*(x^*, t^*)$. Therefore, by choosing α_k to be the magnitude of Q at $x = x^*$ and $t = t^*$, the marginal sensitivity of the performance measure is equal to the adjoint state. In other words, the adjoint state describes the sensitivity of the performance measure to a unit source at any location in the space-time domain.

3.2. Complete Adjoint Equations

The general adjoint equations that were derived in the previous section (25)–(27) are used to obtain expressions for the adjoint states of resident concentration. The backward-in-time location and travel time probabilities are two possible adjoint states. To use the general equation, we need to define the performance measure P in terms of the functional, $h(\alpha, C)$, as shown in (13). As we will show, to obtain location probability as the adjoint state, we define the performance measure to be resident concentration at a point in the space-time domain, and for travel time probability, we use flux concentration at a point in the space-time domain.

To complete the adjoint equation, we also need to specify the boundary and initial conditions on the state variable C. The boundary conditions depend on the system domain (e.g., semi-infinite domain). From the boundary and initial conditions on C, we can obtain their counterparts for the state sensitivity, ψ; through these conditions, we obtain the boundary and final conditions for the adjoint equation.

The governing equation for contaminant transport in a one-dimensional system in a semi-infinite domain ($-\infty < x \leq 0$) that is bounded at the downgradient boundary by a pumping well is

$$-\frac{\partial C^r}{\partial t} + \frac{\partial}{\partial x}\left(D\,\frac{\partial C^r}{\partial x}\right) - \frac{\partial(vC^r)}{\partial x} + Q(x, t) = 0, \quad (28)$$

where C^r is resident concentration. The appropriate initial and boundary conditions for this equation are

$$C^r(x, t) \rightarrow 0 \qquad x \rightarrow -\infty$$

$$\frac{\partial C^r}{\partial x} = 0 \qquad x = 0$$

$$C^r(x, 0) = 0.$$

Taking the derivative of these equations with respect to an arbitrary parameter α_k gives

$$-\frac{\partial \psi}{\partial t} + \frac{\partial}{\partial x}\left(D\,\frac{\partial \psi}{\partial x}\right) - \frac{\partial(v\psi)}{\partial x} + \frac{\partial}{\partial x}\left(\frac{\partial D}{\partial \alpha_k}\,\frac{\partial C^r}{\partial x}\right) - \frac{\partial}{\partial x}\left(C^r\,\frac{\partial v}{\partial \alpha_k}\right) + \frac{\partial Q(x, t)}{\partial \alpha_k} = 0 \quad (29)$$

$$\psi(x, t) \rightarrow 0 \qquad x \rightarrow -\infty$$

$$\frac{\partial \psi}{\partial x} = 0 \qquad x = 0$$

$$\psi(x, 0) = 0.$$

Using these boundary and initial conditions with (26) and (27), we can obtain the appropriate boundary and final conditions for the adjoint state. Substituting $\psi(x, t) \rightarrow 0$ as $x \rightarrow -\infty$ and $\partial\psi/\partial x = 0$ at $x = 0$ into (26), we are left with

$$-D\psi\,\frac{\partial \psi^*}{\partial x}\bigg|_{x=0} - v\psi^*\psi|_{x=0} - D\psi^*\,\frac{\partial \psi}{\partial x}\bigg|_{x\rightarrow-\infty} = 0. \quad (30)$$

This equation is satisfied if we set $\psi^*(x, t) \rightarrow 0$ as $x \rightarrow -\infty$ and $D\partial\psi^*/\partial x + v\psi^* = 0$ at $x = 0$. These are the boundary conditions for the adjoint equation. Note that the forward problem had a second-type boundary condition at the well, while the adjoint problem has a third-type boundary condition for the well. For the final condition, we substitute initial condition $\psi(x, 0) = 0$ into (27), resulting in

$$\psi^*\psi|_T = 0. \quad (31)$$

For this equation to be satisfied, the final condition for the adjoint equation must be $\psi^*(x, T) = 0$.

The only remaining undefined item in the adjoint equation is the functional $h(\alpha, C^r)$. Here we have indicated the dependence of h on resident concentration, C^r (instead of just C), because we wrote the governing ADE (28) in terms of resident concentration. The form of $h(\alpha, C^r)$ depends on whether we are looking for location probability or travel time probability.

3.2.1. Location probability. Recall that the normalized distribution of resident concentration C^r is equivalent to a probability density function for the location of a solute parcel from a given source. Define the performance measure P as the resident concentration at a point, (x', t'), in the space-time domain, which would usually be the location and time of detection. The appropriate functional h is

$$h(\alpha, C^r) = C^r(x, t)\delta(x - x')\delta(t - t'). \quad (32)$$

Substituting (32) into (13) and integrating over the (x, t) domain gives $P = C^r(x', t')$. For the adjoint equation (25), we need $\partial h/\partial C^r$, which is a Fréchet derivative [*Saaty*, 1981] of $h(\alpha, C^r)$ with respect to the function C^r. Taking the Fréchet derivative of both sides of (32), we obtain (see Appendix D).

$$\frac{\partial h(\alpha, C^r)}{\partial C^r} = \delta(x - x')\delta(t - t'). \quad (33)$$

Note that final time T is an arbitrary upper limit of the time domain in the forward problem. If the performance measure for the forward model is resident concentration at (x', t'), we are only interested in the solution for $t \leq t'$. Thus the upper limit of the time domain in the forward problem can be arbitrarily set to $T = t'$. For the backward problem (see section 2.2), the backward time τ is given by $\tau = T - t = t' - t$. Using $T = t'$ and v and D constant, the adjoint equation (25) and its boundary and final conditions for location probability in a semi-infinite domain are given by

$$-\frac{\partial \psi^*}{\partial t} - D\,\frac{\partial^2 \psi^*}{\partial x^2} - v\,\frac{\partial \psi^*}{\partial x} = \delta(x - x')\delta(t - T), \quad (34)$$

$$\psi^*(x, t) \rightarrow 0 \qquad x \rightarrow -\infty$$

$$D\,\frac{\partial \psi^*}{\partial x} + v\psi^* = 0 \qquad x = 0$$

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

This equation can be solved using Laplace transforms. The solution (derived in Appendix A) is

$$\psi^*(x, t) = \frac{1}{\sqrt{4\pi D(T - t)}}\exp\left\{-\frac{[x - x' + v(T - t)]^2}{4D(T - t)}\right\} \cdot \left[1 + \exp\left\{\frac{-xx'}{D(T - t)}\right\}\right] - \frac{v}{2D}\exp\left\{-\frac{vx}{D}\right\} \cdot \operatorname{erfc}\left(-\frac{x + x' - v(T - t)}{\sqrt{4D(T - t)}}\right) \quad (35)$$

for $-\infty < x \leq 0$ and $0 < t \leq T$. This equation is equivalent to the backward-in-time location probability proposed by *Wilson and Liu* [1995] for a detection at the pumping well (equation (10), where $x' = x_d = 0$ and $\tau = T - t$).

As stated earlier, the adjoint state describes the sensitivity of the performance measure to a unit source at any location in the space-time domain. By this definition, the adjoint state in (35) is the sensitivity of the resident concentration at x' to a source at any other location. Not only is resident concentration related to location probability [*Jury and Roth*, 1990], but in this case of a unit source, they are the same. Thus the adjoint state (35) is location probability.

3.2.2. Travel time probability. Recall that the normalized mass flux vC^f is equivalent to the travel time probability for a given source location. Define the performance measure P as the mass flux at a point (x', t') in the space-time domain. The appropriate functional h is

$$h(\alpha, C^r) = vC^f(x, t)\delta(x - x')\delta(t - t'). \quad (36)$$

Substituting (36) into (13) and integrating over the (x, t) domain gives $P = vC^f(x', t')$. For the adjoint equation (25), we need $\partial h/\partial C^r$. Flux concentration is defined in terms of resident concentration, as shown in (5). Substituting this expression in the functional h, we obtain

$$h(\alpha, C^r) = \left[vC^r - D\frac{\partial C^r}{\partial x}\right]\delta(x - x')\delta(t - t'). \quad (37)$$

Using this equation to solve for the Fréchet derivative, $\partial h/\partial C^r$, gives

$$\frac{\partial h(\alpha, C^r)}{\partial C^r} = v\delta(x - x')\delta(t - t') + D\delta'(x - x')\delta(t - t'), \quad (38)$$

where $\delta'(x)$ is the derivative with respect to x of the Dirac delta function. The second term is derived in Appendix B.

By the argument used in the location probability problem, we can arbitrarily take $T = t'$. Thus, for travel time probability in a semi-infinite domain, with v and D constant, the adjoint equation and its boundary and final conditions are given by

$$-\frac{\partial \psi^*}{\partial t} - D\frac{\partial^2 \psi^*}{\partial x^2} - v\frac{\partial \psi^*}{\partial x} = v\delta(x - x')\delta(t - T) + D\delta'(x - x')\delta(t - T), \quad (39)$$

$$\psi^*(x, t) \to 0 \qquad x \to -\infty$$

$$D\frac{\partial \psi^*}{\partial x} + v\psi^* = 0 \qquad x = 0$$

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

This equation can be solved using Laplace transforms. The solution (derived in Appendix C) is

$$\psi^*(x, t) = -\frac{x - x' - v(T - t)}{4\sqrt{\pi D(T - t)^3}} \cdot \exp\left\{-\frac{[x - x' + v(T - t)]^2}{4D(T - t)}\right\} + \frac{x + x' + 3v(T - t)}{4\sqrt{\pi D(T - t)^3}}\exp\left\{\frac{-xx'}{D(T - t)}\right\} \cdot \exp\left\{-\frac{[x - x' + v(T - t)]^2}{4D(T - t)}\right\} - \frac{v^2}{2D}\exp\left\{\frac{-vx}{D}\right\} \cdot \operatorname{erfc}\left(\frac{-x - x' + v(T - t)}{\sqrt{4D(T - t)}}\right) \quad (40)$$

for $-\infty < x \leq 0$ and $0 < t \leq T$. This equation is equivalent to the backward-in-time travel time probability proposed by *Wilson and Liu* [1995] for a detection at the pumping well (equation (12), with $x' = x_d = 0$ and $\tau = T - t$).

The adjoint state in (40) is the sensitivity of the mass flux at x', e.g., the detection location, to a unit source at any other location. Not only is mass flux related to travel time probability [*Jury*, 1982], but in this case, they are the same. Thus the adjoint state (40) is travel time probability.

4. Conclusions

Backward-in-time location and travel time probabilities can be developed heuristically from the forward-in-time resident and flux concentration distributions, as *Wilson and Liu* [1995] showed for a one-dimensional, semi-infinite domain. Although they arrived at the appropriate governing equation and boundary conditions, their approach was based more on intuition than on proof. To provide a consistent framework for obtaining backward-in-time probabilities for multidimensional problems and all spatial domains, we propose the adjoint method.

In this paper, we demonstrated that the adjoint method provides a formal framework for obtaining these backward-in-time probabilities. We derived the one-dimensional adjoint equation for backward-in-time location and travel time probabilities in terms of general boundary conditions. By applying the general form (25)–(27) of the equation to the special case of a semi-infinite domain, we derived expressions for location and travel time probabilities; then we verified that these probabilities are equivalent to those obtained by *Wilson and Liu* [1995]. The general form of the adjoint equation can be used to find location and travel time probabilities for other boundary conditions.

Backward-in-time probabilities can be used to obtain information about where contamination was located before it was detected. They have a variety of applications, including capture zone delineation. The backward model is more efficient than the forward model in situations in which the number of known or potential sources is greater than the number of detections. The benefit of the backward model is that, for each detection, we solve the adjoint equation only once to obtain the backward-in-time location probability for all prior locations at a given time. In other words, for each detection, we obtain information about all possible prior locations after solving the adjoint equation only once. With the forward model, we obtain information about all possible future locations for contamination that was injected at one specified source. Thus, if we have a few detections and many known or possible source locations, the backward model is computationally more efficient than the forward model in that fewer simulations must be run (i.e., one simulation for each detection). However, if we have many detections and only a few possible source locations, the forward model is more computationally efficient than the backward model.

The approach described in this paper is for a simple, idealistic, one-dimensional system. By using the adjoint method, development of the backward probabilities can now formally be extended to a multidimensional system.

Appendix A: Derivation of the Solution to the Adjoint Equation for Location Probability

The adjoint equation for location probability is shown in (34). An equivalent equation is given here, with a new time variable, $\tau = T - t$, and a new space variable, $y = -x$.

$$\frac{\partial \psi^*}{\partial \tau} - D\frac{\partial^2 \psi^*}{\partial y^2} + v\frac{\partial \psi^*}{\partial y} = \delta(y-y')\delta(\tau), \quad (41)$$

$$\psi^*(y,\tau)\to 0 \qquad y\to\infty$$

$$-D\frac{\partial \psi^*}{\partial y} + v\psi^* = 0 \qquad y=0$$

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

where $y' = -x'$ and $0 \le y < \infty$.

Taking the Laplace transform with respect to time ($\tau \to s$) gives

$$s\Psi - \psi^*(y,0) - D\frac{d^2\Psi}{dy^2} + v\frac{d\Psi}{dy} = g(y) \quad (42)$$

$$\Psi \to 0 \qquad y\to\infty$$

$$v\Psi - D\frac{d\Psi}{dy} = 0 \qquad y=0,$$

where Ψ is the transformed state of ψ^* and $g(y) = \delta(y-y')$. The second term on the left-hand side is equal to zero, according to the initial condition in τ (final condition in t). Taking a second Laplace transform with respect to y ($y \to r$) gives

$$s\hat{\Psi} - Dr^2\hat{\Psi} + Dr\Psi|_{y=0} + D\left.\frac{d\Psi}{dy}\right|_{y=0} + vr\hat{\Psi} - v\Psi|_{y=0} = \hat{g}(r) \quad (43)$$

where $\hat{\Psi}$ is the transformed state of Ψ and $\hat{g}(r)$ is the transform of $g(y)$. The fourth and sixth terms on the left-hand side sum to zero according to the boundary condition at $y = 0$. This equation can be rearranged to give

$$\hat{\Psi} = \frac{\hat{g}(r) - Dr\Psi|_{y=0}}{-Dr^2 + vr + s}. \quad (44)$$

Taking the inverse Laplace transform with respect to r ($r \to y$) gives

$$\Psi = L^{-1}_{r\to y}\left[\frac{\hat{g}(r) - Dr\Psi|_{y=0}}{-D(r^2 - vr/D - s/D)}\right] \quad (45)$$

where L^{-1} denotes the inverse Laplace transform.

Using partial fractions, the convolution theorem, and applying the boundary condition as $y \to \infty$, we obtain the following expression for Ψ:

$$\Psi = \frac{1}{v\xi}\left[\exp\left\{\frac{vy}{2D}(1+\xi)\right\} \cdot \int_y^\infty g(y'')\exp\left\{-\frac{vy''}{2D}(1+\xi)\right\}dy'' + \exp\left\{\frac{vy}{2D}(1-\xi)\right\} \cdot \left(\int_0^y g(y'')\exp\left\{-\frac{vy''}{2D}(1-\xi)\right\}dy'' - \frac{1-\xi}{1+\xi}\int_0^\infty g(y'')\exp\left\{-\frac{vy''}{2D}(1+\xi)\right\}dy''\right)\right], \quad (46)$$

where $\xi = \sqrt{1 + 4sD/v^2}$.

The adjoint state ψ^* is found by taking the inverse Laplace transform of the previous equation with respect to s. After some algebraic manipulation and use of the shifting property, we obtain

$$\psi^* = \frac{1}{\sqrt{4\pi D\tau}}\int_0^\infty g(y'')\exp\left\{-\frac{(y-y''-v\tau)^2}{4D\tau}\right\}dy'' + \frac{1}{\sqrt{4\pi D\tau}}\int_0^\infty g(y'')\exp\left\{\frac{-yy''}{D\tau}\right\} \cdot \exp\left\{-\frac{(y-y''-v\tau)^2}{4D\tau}\right\}dy'' - \frac{v}{2D}\exp\left\{\frac{vy}{D}\right\}\int_0^\infty g(y'')\,\mathrm{erfc}\left(\frac{y+y''+v\tau}{\sqrt{4D\tau}}\right)dy''. \quad (47)$$

Recall that $g(y'') = \delta(y'' - y')$. Substituting this expression into (47), evaluating the integrals, and substituting $y = -x$, results in the expression for the adjoint state shown in (35).

Appendix B: Verification of the Equality Used in the Travel Time Probability Adjoint Equation

In the derivation of the adjoint equations (section 3.2), we made use of the following weak equality:

$$\frac{\partial}{\partial C^r}\left[-D\frac{\partial C^r}{\partial x}\delta(x-x')\delta(t-t')\right] = D\delta'(x-x')\delta(t-t'). \quad (48)$$

We justify this substitution here.

By taking the Fréchet derivative [*Saaty*, 1981] with respect to C^r of the bracketed term in (48), we obtain the following operator, L:

$$L = -D\delta(x-x')\delta(t-t')\frac{\partial}{\partial x}. \quad (49)$$

Two operators are weakly equal if the results of their operation on a test function are equal, i.e. $L_1 = L_2$ if $\langle L_1, \phi\rangle = \langle L_2, \phi\rangle$, where ϕ is an arbitrary test function, and $\langle \eta, \xi\rangle$ represents the inner product. For the operator shown in (49), we have

$$\langle L, \phi\rangle = \iint_{x,t} -D\delta(x-x')\delta(t-t')\frac{\partial\phi}{\partial x}\,dx\,dt, \quad (50)$$

where integration is over the entire space-time domain. Integrating the right-hand side by parts in x, we see that

$$\langle L, \phi\rangle = \iint_{x,t} D\delta'(x-x')\delta(t-t')\phi\,dx\,dt. \quad (51)$$

This can be written as

$$\langle L, \phi\rangle = \langle D\delta'(x-x')\delta(t-t'), \phi\rangle. \quad (52)$$

Therefore $L = D\delta'(x-x')\delta(t-t')$.

Appendix C: Derivation of the Solution to the Adjoint Equation for Travel Time Probability

The adjoint equation for travel time probability is shown in (39). An equivalent equation is given here, with a new time variable, $\tau = T - t$, and a new space variable, $y = -x$.

$$\frac{\partial \psi^*}{\partial \tau} - D\frac{\partial^2 \psi^*}{\partial y^2} + v\frac{\partial \psi^*}{\partial y} = v\delta(y-y')\delta(\tau) - D\delta'(y-y')\delta(\tau), \qquad (53)$$

$$\psi^*(y, \tau) \rightarrow 0 \qquad y \rightarrow \infty$$

$$-D\frac{\partial \psi^*}{\partial y} + v\psi^* = 0 \qquad y = 0$$

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

where $y' = -x'$ and $0 \leq y < \infty$.

Taking the Laplace transform of the previous equation with respect to time ($\tau \rightarrow s$) gives

$$s\Psi - \psi^*(y, 0) - D\frac{d^2\Psi}{dy^2} + v\frac{d\Psi}{dy} = g(y) \qquad (54)$$

$$\Psi \rightarrow 0 \qquad y \rightarrow \infty$$

$$v\Psi - D\frac{d\Psi}{dy} = 0 \qquad y = 0,$$

where Ψ is the transformed state of ψ^* and $g(y) = v\delta(y - y') - D\delta'(y - y')$. The second term on the left-hand side is equal to zero, by the initial condition in τ. With the right-hand side written in general terms, (54) is equivalent to (42). Therefore the solution to (53) in general form is (47). Substituting the expression for $g(y)$ into (47), evaluating the integrals, and substituting $y = -x$ gives the expression for the adjoint state shown in (40).

Appendix D: Fréchet Derivative

The derivative, $\partial h/\partial C^r$, in (14) and (25), is a Fréchet (strong) derivative of a functional, h [*Saaty*, 1981; *Zwillinger*, 1989]. Consider a function $u(x)$, where x is space, and a functional $w(u) = Wu$, where W is an operator. The Fréchet derivative, $\partial w/\partial u$, is defined as [*Zwillinger*, 1989]

$$\lim_{\|\varepsilon\|\rightarrow 0} \frac{\|W[u+\varepsilon] - Wu - \varepsilon Lu\|}{\|\varepsilon\|} = 0, \qquad (55)$$

where L the derivative operator, Lu is the Fréchet derivative ($Lu = \partial w/\partial u$), and $\|\cdot\|$ represents the norm. If, for example, $w(u) = Wu = u^3 + u'' + (u')^2$, then the derivative operator is $L[\ ] = 3u^2[\ ] + [\ ]'' + 2u'[\ ]'$. Contrast this to the derivative of a function; in this example, $\partial w/\partial x = 3u^2u' + u''' + 2u'u''$. In our adjoint problem for location probability, the functional is $h(\alpha, C^r) = C^r(x, t)\delta(x - x')\delta(t - t')$, and its operator is $H[\ ] = [\ ]\delta(x - x')\delta(t-t')$. Rewriting (55) to define the derivative of this functional gives

$$\lim_{\|\Delta C^r\|\rightarrow 0} \frac{\|H[C^r + \Delta C^r] - HC^r - \Delta C^r LC^r\|}{\|\Delta C^r\|} = \lim_{\|\Delta C^r\|\rightarrow 0} \frac{\|\Delta C^r\delta(x-x')\delta(t-t') - \Delta C^r LC^r\|}{\|\Delta C^r\|} = 0, \qquad (56)$$

so that $\partial h/\partial C^r = LC^r = \delta(x - x')\delta(t - t')$. By a similar process, we can show that (38) is the Fréchet derivative of (37).

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