Analysis of tracer tomography using temporal moments of tracer breakthrough curves

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A B S T R A C T

Hydraulic/partitioning tracer tomography (HPTT) was recently developed by Yeh and Zhu [Yeh T-C, Zhu J. Hydraulic/partitioning tracer tomography for characterization of dense nonaqueous phase liquid source zones, Water Resour Res 2007;43:W06435. doi:10.1029/2006WR004877.] for estimating spatial distribution of dense nonaqueous phase liquids (DNAPLs) in the subsurface. Since discrete tracer concentration data are directly utilized for the estimation of DNAPLs, this approach solves the hyperbolic convection–dispersion equation. Solution to the convection–dispersion equation however demands fine temporal and spatial discretization, resulting in high computational cost for an HPTT analysis. In this work, we use temporal moments of tracer breakthrough curves instead of discrete concentration data to estimate DNAPL distribution. This approach solves time independent partial differential equations of the temporal moments, and therefore avoids solving the convection–dispersion equation using a time marching scheme, resulting in a dramatic reduction of computational cost. To reduce numerical oscillations associated with convection dominated transport problems such as in inter-well tracer tests, the approach uses a finite element solver adopting the streamline upwind Petrov–Galerkin method to calculate moments and sensitivities. We test the temporal moment approach through numerical simulations. Comparing the computational costs between utilizing moments and discrete concentrations, we find that temporal moments significantly reduce the computation time. We also find that tracer moment data collected through a tomographic survey alone are able to yield reasonable estimates of hydraulic conductivity, as indicated by a correlation of 0.588 between estimated and true hydraulic conductivity fields in the synthetic case study.

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1. Introduction

Effective remediation of sites contaminated with dense nonaqueous phase liquids (DNAPLs) relies on adequate characterization of the size, nature, and distribution of DNAPL source zones [23]. Because of the various human activities of the contaminated sites and complex subsurface environment, characterization of DNAPL source zones is a difficult task. Since the 1980s, a variety of methods have been developed and applied to the DNAPL source zone characterization. Some of the methods yield localized point measurements of DNAPLs (i.e., core retrieval and analysis) at a limited number of locations, while others produce average saturation over a relatively large volume of geologic medium (i.e., partitioning tracer tests) [23]. Very few techniques have been developed to estimate detailed spatial distribution of DNAPL saturation in the subsurface.

The concept of tomography, originally developed in medical sciences for detailed imaging of a human body (for example, computerized axial tomography or CAT), has been recently introduced into subsurface hydrology. Tomographic surveys allow one to image an object from different angles and perspectives, and therefore provide high resolution images of the object. Utilizing the tomography concept, hydrologists have developed the hydraulic tomography (HT) technique to estimate detailed spatial variation of hydraulic properties of the subsurface [25], Butler and Liu [3], [6,22,29,31,33,36,37] and others. Specifically, in hydraulic tomography, a series of aquifer tests are sequentially conducted at different locations with head responses being collected from other locations during each test. Head responses from all the tests are then used in an inverse method to estimate hydraulic parameters. Results from sandbox experiments by Liu et al. [20], Illman et al. [9,10], and Liu et al. [19] showed that transient HT can identify not only the pattern of the heterogeneous hydraulic conductivity (K) field, but also the variation of specific storage (Ss) in the sandbox. Moreover, these estimated K and Ss fields from the HT sandbox...
experiments accurately predicted the drawdown evolution caused by a pumping test that was not used in the HT analysis. Likewise, a recent application of HT to a well field at Montalto Uffugo Scalo, Italy, produced an estimated transmissivity field that is deemed to be consistent with the geology of the site [26]. Bohling et al. [1] and Li et al. [17,18] also showed promising results of HT in their field experiments.

Success of HT has led Yeh and Zhu [34] to the development of a joint hydraulic and tracer tomography for characterizing of DNAPL source zones in the subsurface. Yeh and Zhu [34] advocated that migration and distribution of DNAPLs are highly controlled by hydraulic heterogeneity of the subsurface. HT delivers high resolution imaging of the hydraulic heterogeneity which controls tracer migration path and thus the likely locations of DNAPLs. Conducting partitioning tracer tests in a tomographic fashion afterwards can facilitate detailed delineation of the spatial distribution of DNAPLs.

The proposed joint hydraulic/tracer tomography survey consists of a series of steady state water injection and tracer tests in the DNAPL source zones at different locations, and monitoring head changes and tracer breakthrough curves (BTCs) at many observations locations. Using the sequential successive linear estimator, the head responses are used first to provide detailed estimates of hydraulic conductivity spatial variation, and the BTCs from tracer tomography are then used to estimate spatial variations of water content and DNAPL content. Notice that conducting multiple tracer tests in the field is practically challenging due to cost and time. Yeh and Zhu [34] on the other hand argued that one or two dual tracer tests are sufficient to estimate DNAL distribution when the usefulness of hydraulic tomography is maximized.

Generally speaking, estimating DNAPL spatial variation using discrete tracer concentration data in partitioning tracer tests imposes significant computational cost (e.g. [34, 35]). This computational burden arises from the requirement of fine spatial and temporal discretizations of the advection–dispersion equation, which is solved repeatedly during the estimation procedure. Instead of using discrete concentration values at different times, many researchers have used temporal moments of tracer BTCs to estimate hydraulic properties or DNAPL content in the subsurface. Approaches based on the temporal moments solve equations that are functions of spatial coordinates only. Thus the approaches avoid time marching procedures and offer computational advantages over those based on the discrete concentration values. For instance, Jin et al. [14] used temporal moments of BTCs to estimate the average DNAPL saturation for 1D and 2D problems. Harvey and Gorelick [7] developed temporal moment-generating equations that directly solve temporal moments without solving the entire BTCs. Using the generating equations, James et al. [11] calculated the covariances of temporal moments and cross covariances between temporal moments and parameters. James et al. [12] further used temporal moments to estimate DNAPL spatial variation. Cirpka and Kitanidis [4] combined hydraulic heads and the first moments of tracer BTCs to infer hydraulic conductivity spatial distribution. Based on Lagrangian conceptualization of flow fields, Jawitz et al. [13] implemented partitioning tracer higher moments to estimate statistical parameters for NAPL source zones. In spite of these applications, the tracer moment approach has not been applied to tracer tomography.

In this work, the tracer temporal moment method is applied to the joint hydraulic/tracer tomography approach. The aim of the application is to reduce the computational cost associated with the joint approach such that the approach can be appealing for practitioners. In this study, we implement tracer temporal moments in sequential successive linear estimator (SSLE) to interpret the joint hydraulic/tracer tomography data for estimating DNAPL spatial distribution. We first present temporal moment equations for the transport of a partitioning tracer. We next apply a stream-line upwind/Petrov–Galerkin formulation to reduce numerical oscillation due to discretization in solving convection dominated moment equations. Then, we incorporate the first temporal moment of tracer into the SSLE inverse algorithm, which requires evaluation of sensitivity of temporal moments to hydraulic conductivity, water content, and DNAPL content, along with calculation of covariance of moments and cross covariance between moments and the above three parameters. Finally, we test the temporal moment method using the same synthetic cases in Yeh and Zhu [34] and compare the computational cost. The results from temporal moments are also compared with those from inversion of discrete concentration data. The usefulness of temporal moments of tracer BTCs collected in a tomographic way in estimating hydraulic conductivity is also investigated.

2. Methodology

2.1. Temporal moment equations for a partitioning tracer

As mentioned previously, the joint hydraulic/tracer tomography survey involves a series of steady state water injection and tracer tests in the DNAPL source zones at different locations, and monitoring head changes and BTCs at many observation locations. Interpreting this joint hydraulic/tracer tomography survey requires both forward and inverse modeling efforts. The forward modeling solves steady state flow equation and tracer temporal moment equations, which are discussed below.

2.1.1. Governing equations

We assume that the steady state flow field induced by each tracer test in the joint hydraulic/tracer tomography is described by

$$\nabla \cdot (K(x) \nabla H) + Q = 0$$  \hspace{1cm} (1)

subject to boundary conditions:

$$H|_{\Gamma_1} = H_1, \quad [K(x) \nabla H] \cdot \mathbf{n}|_{\Gamma_2} = q_b$$  \hspace{1cm} (2)

where $H$ is the total head, $x$ is the spatial coordinates ($x = [x_1, x_2, x_3]$), $L$, and $x_3$ represents the vertical coordinate and is positive upward. The hydraulic conductivity field is denoted by $K(x) (L/T)$. In addition, $Q$ is the source term ($1/T$). In Eq. (2), $H_1$ is the prescribed total head at the Dirichlet boundary $\Gamma_1$, $q_b$ is the specific flux ($L/T$) at the Neumann boundary $\Gamma_2$, and $\mathbf{n}$ is a unit vector normal to the boundary.

The transport of a partitioning tracer in the joint hydraulic/tracer tomography is assumed to be governed by the advection–dispersion–retardation equation:

$$\frac{\partial}{\partial t}(\theta_a c + \theta_h c_p) = -\nabla \cdot (\mathbf{q} c) + \nabla \cdot (\theta_a \mathbf{D} \nabla c) + Q c N(x - x_q)(t - t_s)$$  \hspace{1cm} (3)

subject to following initial and boundary conditions:

$$C_{c=0} = c_0, \quad \left. C \right|_{\Gamma_1} = c_1, \quad \mathbf{q} c = -\theta_a \mathbf{D} \nabla C|_{\Gamma_2} = q_i$$  \hspace{1cm} (4)

where $c$ is the partitioning tracer concentration ($M/L^3$); $\theta_a$ is the volumetric water content and $\theta_h$ is the volumetric DNAPL content. Notice that porosity $\theta$ is the sum of $\theta_a$ and $\theta_h$. $K_p$ is the partitioning coefficient, $\mathbf{q}$ is Darcy velocity vector ($L/T$), which is assumed to be time invariant and is calculated from Eqs. (1) and (2); $\mathbf{D}$ is dispersion tensor and is given as

$$D_i = \frac{\nu_i D_i}{|\mathbf{V}|} (x_i - x_t) + \delta_{ij}(x_{i'j'} V + D_j')$$  \hspace{1cm} (5)

where $x_i$ and $x_t$ are the longitudinal and transverse dispersivities ($L$), respectively; $\nu_i$ is the seepage velocity ($L/T$) defined as $q_i/\theta_w$; $|\mathbf{V}|$ is the magnitude of the seepage velocity; $\delta_{ij}$ is the Kronecker
delta function which equals unity if the indices are identical and zero otherwise, and \( D_J \) is the molecular diffusion coefficient (\( L^2/T \)). In addition, \( c_i \) is the concentration of the injecting tracer solution; \( N(x-x_0)(t-t_0) \) denotes the product of a unit impulse function in space (equals unity only at location \( x_0 \)) and a step function in time (equals unity from \( t = 0 \) until \( t = t_0 \) and zero afterwards), where \( t_0 \) denotes the time when the release of partitioning tracer is terminated; \( C_0 \) is the initial tracer concentration, which is typically zero for a tracer test; \( C_1 \) is the prescribed concentration at the Dirichlet boundary \( I_1 \); \( q_i \) is tracer flux at the Neumann boundary \( I_2 \).

2.2. Temporal moment equations for a tracer BTC

The \( k \)th temporal moments of a tracer BTC at location \( x \) can be defined as

\[
m_k = \int_{t=0}^{t=\infty} t^k c(x, t) dt
\]

where \( k \) is the order of moment. Multiplying Eq. (3) by \( t^k \), integrating over time, assuming \( q \) is time invariant, and substituting Eq. (6) into Eq. (3) yield moment-generating equation

\[
(\theta_w + K_0 \theta_h) t^k c_{t=0} - (\theta_w + K_0 \theta_h) m_{k-1} = -\nabla \cdot (\theta_w \nabla m_k) + \nabla \cdot (\theta_w \nabla m_k) + Q c_N(x-x_0)(t-t_0) \int_{t=0}^{t=\infty} t^k dt
\]

For a finite pulse injection tracer test, the initial concentration is commonly zero and the final concentration is also zero after the entire BTC passes. Therefore, the first term on the left hand side disappears. Then, Eq. (7) is simplified to James et al. [12] and Cirpka and Kitanidis [4]

\[
\nabla \cdot (\theta_w \nabla m_k) - \nabla \cdot (\theta_w \nabla m_k) - Q c_N(x-x_0)(t-t_0) \int_{t=0}^{t=\infty} t^k dt = (\theta_w + K_0 \theta_h) m_{k-1}
\]

subject to the boundary conditions:

\[
m_k|_{I_1} = \int_{t=0}^{t=\infty} t^k c_{t=0} dt, \quad m_0|_{I_2} = \int_{t=0}^{t=\infty} t^k q_i dt
\]

Notice that the temporal moment equation (8) eliminates time dependence in Eq. (3). In theory, Eqs. (8) and (9) can be applied to temporal moments of any order. However, our study will focus only on the zeroth moment \( m_0 \) (mass under the BTC) and first moment \( m_1 \). Notice that \( m_1/m_0 \) is mean arrival time of the BTC. Setting \( k = 0 \) gives the zeroth moment equation,

\[
\nabla \cdot (\theta_w \nabla m_0) - \nabla \cdot (\theta_w \nabla m_0) - Q c_N(x-x_0)(t-t_0) \int_{t=0}^{t=\infty} t^0 dt = 0
\]

Setting \( k = 1 \) gives the first moment equation,

\[
\nabla \cdot (\theta_w \nabla m_1) - \nabla \cdot (\theta_w \nabla m_1) - Q c_N(x-x_0)(t-t_0) \int_{t=0}^{t=\infty} t dt = (\theta_w + K_0 \theta_h) m_0
\]

In order to obtain the first moment, Eq. (10) must be solved first for the zeroth moment and the result is then used in Eq. (11) to solve for the first moment.

2.2. Numerical method for moment equations

The moment-generating equation (8) is in the form of steady state convection dispersion equation. Application of the traditional Galerkin finite element method to the moment equations can suffer from oscillations if these equations are convection dominated (i.e., Pelet number is higher than 1). To reduce the oscillations, we use a Streamline-Upwind Petrov–Galerkin (SUPG) method which in principle adds an artificial dispersion (also called stabilization factor) to the streamline direction of the velocity field [2]. More specifically, the weighting function of the finite element method is perturbed as follows:

\[
\tau = N_i + \tau' q \cdot \nabla N_i
\]

where \( N_i \) is the shape function used in the finite element method to solve tracer moment equations. There are several choices of the value of \( \tau' \) and we use the suggestion from [12], namely

\[
\tau' = \frac{\eta q L_x + \eta q L_y + \eta q L_z}{2(q^2 + q_y^2 + q_z^2)}
\]

where \( L_x, L_y, \) and \( L_z \) denote the element length in \( x, y, \) and \( z \)-directions, respectively. Moreover \( \tilde{\varepsilon} \) is defined as

\[
\tilde{\varepsilon} = \begin{cases} \frac{\bar{\varepsilon}}{2} & \text{if } |x_c| < 0.5 \varepsilon \text{ otherwise} \end{cases}
\]

where \( \bar{\varepsilon} \) and \( \varepsilon \) are defined similarly.

For implementation of the convection dispersion solver, we use the Diffpack framework [15], which is a collection of object-oriented C++ libraries for solving partial differential equations. In addition to different stabilization techniques, such as those defined in Eqs. (12)–(14), Diffpack also has an extensive collection of different element types and iterative solvers of linear systems. The resulting convection dispersion solver is programmed as an efficient and flexible plug-in, which fits into an in-house FORTRAN code for hydraulic/tracer tomography.

2.3. Inverse method

2.3.1. Implementation of temporal moments of tracer in SSLE inverse method

The sequential successive linear estimator (SSLE) is a geostatistical inverse approach that conceptualizes the parameter fields to be estimated as spatial stochastic processes, and seeks effective parameter fields conditioned on available state information (i.e., measurements of aquifer responses) and parameter measurements. The estimator utilizes statistical moments of parameter fields (i.e., mean and variance) as a priori information and uses partial differential equations of physical processes to relate state information to parameter fields. The SSLE has been extensively discussed and applied to a variety of inverse problems in hydrogeology and geophysics. Here, we will only briefly present the SSLE method.

Similar to Yeh and Zhu [34], the natural logarithms of \( K, \theta_w, \) and \( \theta_h \) fields are treated as stochastic processes, \( Y_i \) (i.e., \( Y_1 = \ln K; \ Y_2 = \ln \theta_w; \ Y_3 = \ln \theta_h \)), which are represented by a mean \( Y_i \) and perturbation \( Y_i \) component, such that \( Y_i = Y_i + Y_i \). Likewise, the first tracer moment is conceptualized as a stochastic process with a mean \( m_1 \) and a perturbation \( \varepsilon \), such that \( m_1 = m_1 + \varepsilon \). Using the logarithms of \( \theta_w \) and \( \theta_h \), instead of using \( \theta_w \) and \( \theta_h \) themselves, is merely to avoid producing estimates with negative values. As a result, it is impossible to produce an estimated DNAPL content value of \( \theta_h = 0 \), which is undesirable since \( \theta_h = 0 \) exists in many field conditions. It is practical that a very small value of estimated \( \theta_h \) could be treated as an indication of absence of DNAPL. Note that [4] reported that the sensitivities of the zeroth moment to parameters were essentially zero, resulting in very limited information about the spatial variability of parameters. As a result, we will focus on using the first moment for the DNAPL estimation.

The SSLE starts with the classic colriging method which uses direct measurements of the parameter and measurements of the first
moment at some sample locations to obtain a linear estimate of the parameters $y_l$ in the entire domain, that is
\[
\hat{y}_l = αy'_l + βe'
\]
where $Y'_l$ and $e'$ are $N_p \times 1$ and $N_e \times 1$ vectors of measurements of parameters and measurements of the first moments, respectively; $N_p$ is the number of parameter measurements and $N_e$ is number of the first moment measurements obtained from one tracer test; $α$ and $β$ are $N_p \times N$ and $N_e \times N$ weight matrices and $N$ is the total number of finite element cells in the simulation domain. The weights represent contributions of each measurement to the estimates. The weights are calculated based on unconditional spatial moments (means and covariances) of $Y_l$, the spatial covariances of $m_1$, and spatial cross covariances between $m_1$ and $Y_l$. The statistical moments of $Y_l$ are input parameters. Using the moments of $Y_l$, statistical moments of $m_1$ are subsequently determined by a first order approximation.

Cokriging is a linear estimator. Therefore, it can not fully exploit the usefulness of $m_1$ because the relation between $m_1$ and $Y_l$ is not linear. SSLE uses an iterative approach to include the nonlinear relation and to subsequently improve the estimates. That is, the estimates of parameter fields at $r$th iteration $\hat{y}_l^r$ are used in the forward models (Eqs. (10) and (11)) to approximate the conditional mean of $m_1$. The differences between observed first moments $e'$ and calculated first moments $e^r$ are subsequently used in conjunction with associated weights $ω^r$ to improve the estimate $\hat{y}_l^{r+1}$.
\[
\hat{y}_l^{r+1} = \hat{y}_l^r + ω^r (e' - e^r)
\]
These weights are obtained by solving a system of equations that involve conditional covariances and cross covariances, which are derived from the previous iteration. After improved estimates are obtained, the conditional covariance of $y_l$, $R^e_{y_l}$, is updated through the following equation:
\[
R^e_{y_l} = R^e_{y_l} - ω^r R^e_{y_l}
\]
where $R^e_{y_l}$ denotes the conditional cross covariance between $y_l$ and the $m_1$ measurements at iteration $r$. The updated conditional covariance is then used in the next iteration. The aforementioned steps continue until the improvement in the estimates diminishes to a prescribed value. Specifically, SSLE stops when both the variances of the estimated parameter fields and the differences between the observed and calculated first moments stabilize. SSLE can simultaneously include all the measurements collected from a tracer tomography survey jointly; however, this can lead to numerical and computer memory problems because of an extremely large system of equations [8]. To avoid this problem, the $m_1$ measurements

![Fig. 1. Comparison between true and estimated DNAPL content fields: (a) true field; (b) estimated from discrete concentration data from eight tracer tests; (c) estimated from tracer first moment data from eight tracer tests and (d) estimated from tracer first moment data from one tracer test.](image-url)
collected from a tracer tomography survey are added into the inverse process sequentially, such as one tracer test is followed by another tracer test. By adding secondary information in a sequential way, the procedure of successive improvement as described above is used for every tracer test.

2.3.2. Calculation of sensitivities
As indicated above, to use tracer moments in SSLE, one needs to calculate the covariance of $m_1$ measurements and cross covariances between $m_1$ measurements and $Y_l$. They are evaluated by a first order approximation as described in Yeh and Zhu [34]. The first order approximation method requires the sensitivities of $m_1$ to $Y_l$. While the process of calculating covariance and cross covariance is essentially the same as that in Yeh and Zhu [34], evaluation of sensitivities is different because the equations describing the relation between the $m_1$ to $Y_l$ are different from the equations relating tracer concentrations to $X_e$.

The adjoint state method is most efficient when the number of measurements is far smaller than the number of parameters. Detailed description of adjoint state method can be found in Sykes et al. [28], Sun [27], and Cirpka and Kitanidis [4]. Here, we briefly describe the adjoint state method for calculating sensitivity of $m_1$ to $ln K$, $ln \theta_m$, and $ln \theta_u$. The adjoint state method can be described as three steps. The first step is to solve for mean head $H$ using Eqs. (1) and (2) and to solve mean zeroth and first tracer moments $m_0$ and $m_1$ using Eqs. (10) and (11) with associated boundary conditions. The second step is to solve three adjoint equations for a first moment measurement $m_1$ at location $x_e$ to obtain three adjoint states, $\phi_1, \phi_0$, and $\phi_h$:

$$- q \cdot \nabla \phi_1 - \nabla \cdot (\bar{\theta}_u D \nabla \phi_1) = - \frac{\partial P}{\partial m_1} \quad (18)$$

$$- q \cdot \nabla \phi_0 - \nabla \cdot (\bar{\theta}_u D \nabla \phi_0) = - \frac{\partial P}{\partial m_0} + \phi_1 \bar{\theta}_u + \phi_1 K_N \theta_n \quad (19)$$

$$\nabla \cdot (K \nabla \phi_h) = - \nabla \cdot (K \phi_0 \nabla m_0) - \nabla \cdot (K \phi_1 \nabla m_1) - \frac{\partial P}{\partial H} \quad (20)$$

subject to the following boundary conditions:

$$\phi_0 |_{r_1} = 0, \quad \phi_k |_{r_1} = 0, \quad (\phi_1 q + D \nabla \phi_1) \cdot n |_{r_2} = 0$$

$$(-K \nabla \phi_0 - K \phi_0 \nabla m_0 - K \phi_1 \nabla m_1) \cdot n |_{r_2} = 0 \quad (21)$$

In the above equations $P = m_1 (x - x_e)$ represents the first moment measurement at location $x_e$ and $\delta$ is Kronecker’s delta function, which equals one if $x = x_e$ and zero otherwise; $k$ is the adjoint state index, equals either 0 or 1. The third step is then to calculate the sensitivities of $m_1$ to parameters

$$\frac{\partial m_1}{\partial \ln K_j} = \int_{\Omega_j} (-K \nabla \phi_0 \nabla H - \phi_0 \nabla m_0 K \nabla H - \phi_1 \nabla m_1 K \nabla H) d\Omega \quad (22)$$

$$\frac{\partial m_1}{\partial \ln \theta_m} = \int_{\Omega_j} (\bar{\theta}_u D \nabla \phi_0 \nabla m_0 + \bar{\theta}_u D \nabla \phi_1 \nabla m_1 - \phi_1 \bar{\theta}_u \nabla m_0) d\Omega \quad (23)$$

$$\frac{\partial m_1}{\partial \ln \theta_u} = \int_{\Omega_j} - \phi_1 K_N \theta_u m_0 d\Omega \quad (24)$$

where $\Omega_j$ is the sub-volume of location $j$. In the adjoint state method, Eqs. (1), (2), (11), and (12) only need to be solved once while Eqs. (18)–(21) are solved once for each $m_1$ measurement.

3. Synthetic case study

3.1. Comparison with inversion using tracer concentrations

To test the computational efficiency of the proposed inverse method for tracer tomography, we apply the method to the synthetic quasi-three dimensional confined aquifer that was used by Yeh and Zhu [34]. The synthetic aquifer is 40 m long in horizontal direction, 0.5 m in width, and 10 m in vertical direction. The parameter fields of $K, \theta_m$, and $\theta_u$ in the heterogeneous aquifer are assuming to be lognormal and are generated using a spectral method. The $K$ field has a geometric mean of 0.86 m/d with variance of 1.68 (0.8 for $ln K$); the $\theta_m$ field has a geometric mean of 0.291 with variance of 0.004 (0.057 for $ln \theta_m$); and the $\theta_u$ field has a geometric mean of 0.1 with variance of 0.012 (0.1 for $ln \theta_u$). The longitudinal and transverse dispersivities are constant values of 0.05 m and 0.01 m, respectively. The molecular diffusion is $10^{-7}$ m$^2$/d everywhere. The partitioning coefficient is equal to 9.0. The aquifer is

![Fig. 2. Scatter plot of true DNAPL content field versus estimate fields from: (a) discrete concentration data from eight tracer tests; (b) tracer first moment data from eight tracer tests; (c) tracer first moment data from one tracer test.](Image)
discretized in to 1600 elements with a uniform size of 0.5 m \times 0.5 m \times 0.5 m. The discretization resulted in a Peclet number about 10. Note that we used a synthetic 2D aquifer to demonstrate the method for easy illustration purpose; the method can be readily applied to 3D aquifer systems.

A hydraulic/partitioning tracer survey is applied to the synthetic aquifer. To conduct HPTT, four fully penetrating wells are emplaced in the aquifer. Each well has nine pressure and tracer dual-purpose sampling ports and two injection ports (Fig. 1a). The distance between two adjacent wells is 10 m. The survey consists of eight tracer injection tests. Each test starts with the simulation of a steady state flow field resulting from a continuous injection of water at a rate of 20 m$^3$/d through one of the eight injection ports. Then the migration of a slug of conservative and partitioning tracers is then simulated. The survey collects 288 steady state head measurements, 288 conservative tracer BTCs, and 288 partitioning tracer BTCs from eight tracer injection tests. The first moments can be either calculated from Eq. (6) after entire BTCs are calculated from Eqs. (3) and (4) or directly calculated through solving Eqs. (10) and (11). In reality, the only way to obtain temporal moments is through BTCs, which however are subject to truncation errors [21] as well as noise. As the primary goal of this work is to demonstrate the computational efficiency of employing temporal moments for interpreting BTCs from partitioning tracer tomography, we use Eqs. (10) and (11) to obtain the zeroth and first moments and ignore effects of the potential truncation error on the BTC temporal moment estimates. Also notice that tracer tomography survey employed in this synthetic case is highly idealized and merely serves as the purpose of illustrating the concept of tracer tomography. Collecting 288 conservative tracer BTCs and 288 partitioning tracer BTCs in the field for a domain with this size will be cost-prohibitive.

The process of interpreting hydraulic/partitioning tracer tomography proposed by Yeh and Zhu [34] starts with estimating $K$ using steady state head data collected from hydraulic/tracer tomography; then, conservative tracer concentration data are used to estimate $\theta_w$ and to improve the estimation of $K$. Finally, the partitioning tracer concentration data collected from partitioning tracer tomography are used to estimate $\theta_n$ and to again improve the estimate of $K$. In this work, we follow the same steps of the above interpretation process; but during the last step of the interpretation, we use partitioning tracer first moments instead of discrete concentration data. Two cases were considered. The first case used the first moments from all eight tracer tests and the second case used the first moment from the tracer test initialized at the port located at $x = 24.75$ m and $z = 6.75$ m, where nearby DNAPL core samples show relative higher DNAPL content. The total CPU time for the first case is about 50 min on a single PC (Pentium 4 2.8 GHz), while the CPU time for the inverse modeling of partitioning tracer concentration measurements is about 200 min using an eight node PC cluster with the same processors (2.8 GHz Pentium 4). The saving of computational cost is dramatic. Using the moment approach makes the inverse modeling of the joint hydraulic/tracer tomography feasible using a single PC and thus more attractive for a rapid analysis of the HPTT survey.

Fig. 1 compares the estimated $\theta_n$ field from discrete concentration data (Fig. 1b) with the true field (Fig. 1a) and that (Fig. 1c) using the first moment data from eight tracer tests as well as that (Fig. 1d) using the first moment data from one tracer test. These figures show that, similar to the estimate from discrete concentration data, the estimates from first moments reveal the general pattern of the spatial variation of synthetic $\theta_n$ field but the pattern is smoother than the field estimated from the concentration data. The estimated $\theta_n$ field from the first moment data using one tracer test appears even smoother than that using first moment from eight tracer tests but again captures the general spatial variation of the synthetic $\theta_n$ field. Fig. 2 presents scatterplots of the estimated and true $\theta_n$ fields from these two approaches. The error

![Fig. 3. Comparison between true and estimated hydraulic conductivity fields: (a) true field; (b) estimated from head data; (c) estimated from tracer first moment data from eight tracer tests.](image-url)
The result has not been confirmed for tomographic surveys. Tomography provides significant amount of arrival time information originated from different flow scenarios, which can be effective in estimating hydraulic conductivity in cases where the high density head data are not available. Nevertheless, Cirpka and Kitanidis [4] noticed that using arrival time alone may cause numerical instability during the estimation of the hydraulic conductivity field. To test the usefulness of tracer arrival time data collected in a tomographic survey for estimating K field, the conservative tracer arrival time data were used to estimate K and \( \theta_e \) simultaneously as the conservative tracer arrival time is also affected by \( \theta_w \).

Fig. 3a shows the true hydraulic conductivity field of the synthetic aquifer, which is used to compare with the estimated field based on the head information (Fig. 3b) and that based on the tracer first moment (Fig. 3c). A visual comparison of these figures suggests that the estimated spatial variation of K from head data is a closer approximation of the true in comparison with that from arrival time of conservative tracer. To confirm this finding, scatter plots of the true and estimated fields based on the head and that based on the first moment are illustrated in Fig. 4a and b, respectively, along with the statistical measures (L1, L2, and \( \rho \)). The results substantiate that the estimated K field from the head is clearly superior to that estimated field based on the tracer alone, in agreement with the conclusions by Li and Yeh [16] and Yeh and Zhu [34]. Notice that using partitioning tracer arrival time would involve additional unknowns (\( \theta_e \)) and thus it is expected that it will not yield better results than the conservative tracer arrival times.

3.3. Cross correlation of first temporal moment of tracer to parameters

To understand how tracer temporal moments can be used to estimate different parameters, we calculate the cross correlation of the first temporal moments of partitioning tracer BTCs to selected parameters (i.e., \( K \), \( \theta_e \), and \( \theta_w \)). The cross correlation between the first moment perturbation \( \varepsilon \) at location \( \mathbf{x}_i \) and a parameter perturbation \( y_j \) at location \( \mathbf{x}_j \), \( C_{\varepsilon y}(\mathbf{x}_i, \mathbf{x}_j) \), is calculated through the following equation:

\[
C_{\varepsilon y}(\mathbf{x}_i, \mathbf{x}_j) = \frac{R_{\varepsilon y}(\mathbf{x}_i, \mathbf{x}_j)}{\sqrt{R_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_i)R_{yy}(\mathbf{x}_j, \mathbf{x}_j)}}
\]

where \( R_{\varepsilon y}(\mathbf{x}_i, \mathbf{x}_j) \) is the cross covariance between \( \varepsilon \) and \( y_j \) at locations \( \mathbf{x}_i \) and \( \mathbf{x}_j \); \( R_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_i) \) and \( R_{yy}(\mathbf{x}_j, \mathbf{x}_j) \) are variances of \( \varepsilon \) at \( \mathbf{x}_i \) and \( y_j \) at \( \mathbf{x}_j \), respectively. \( R_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_i) \) and \( R_{yy}(\mathbf{x}_j, \mathbf{x}_j) \) are calculated through a first order approximation [30] as follows:

\[
R_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_m) = \sum_{m=1}^{N} J_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_m) R_{\varepsilon y}(\mathbf{x}_m, \mathbf{x}_j)
\]

\[
R_{y y}(\mathbf{x}_j, \mathbf{x}_m) = \sum_{l=1}^{N} \sum_{m=1}^{N} J_{y y}(\mathbf{x}_j, \mathbf{x}_m) R_{\varepsilon y}(\mathbf{x}_m, \mathbf{x}_j)
\]

where \( J_{\varepsilon \varepsilon}(\mathbf{x}_i, \mathbf{x}_m) \) is the sensitivity of the first moment at location \( \mathbf{x}_i \) with respect to the parameters \( y_j \) at locations \( \mathbf{x}_m \). \( R_{\varepsilon y}(\mathbf{x}_m, \mathbf{x}_j) \) is specified a priori at the first iteration and is evaluated through Eq. (17) at subsequent iterations. SSLE is an iterative estimator and \( R_{\varepsilon \varepsilon} \) and \( R_{y y} \) are reevaluated at each iteration, so are the cross correlations. Here we will examine only the correlation at the first iteration. In this analysis, one of the eight tracer tests in the hydraulic/tracer tomographic survey in the synthetic aquifer is selected for this analysis. The tracer/water injection location is selected at \( x = 14.75 \text{ m}, \ z = 7.25 \text{ m} \) and an observation port is located at \( x = 24.75 \text{ m}, \ z = 4.25 \text{ m} \). The spatial statistics (i.e., mean, correlation scales) for the ‘true’ parameter fields are used for the calculation of the cross correlations, except the variances where are assumed to be the same for all three parameter fields. For comparison, the cross

![Fig. 4. Scatter plot of true hydraulic conductivity field versus estimated field from (a) head data; (b) tracer first moment data from eight tracer tests.](image-url)
correlation between a steady state head at the observation port and $K$ is also calculated.

Fig. 5a–c shows the cross correlations between the perturbation of the first moment of a partitioning tracer at the observation location and perturbations of $\ln K$, $\ln \theta_w$, and $\ln \theta_n$ throughout the aquifer, respectively. In all these figures, the areas with very low correlation values (i.e., the absolute value less than 0.1) are blanked and velocity vectors of the steady state flow induced by the injection are plotted. Note that the velocity vectors are plotted merely for showing the flow direction and are not scaled to the magnitude of flow velocity. These figures demonstrate that the correlation between the first moment and three parameters exhibit similar pattern. That is, the tracer first moment is only correlated to parameters along a narrow strip between the injection location and the BTC measurement location, which follows the water flow line indicated in the figures. The area with absolute correlation value greater than 0.3 between the first moment and $K$ and between the first moment and $\theta_n$ is confined to a small region along the strip. The highest absolute correlation value for $K$ is close to 0.4 and for $\theta_n$ is close to 0.5. On the other hand, the correlation between the first moment and $\theta_w$ is small with the maximum absolute value less than 0.15. Fig. 5d shows the cross correlation between a steady head at the observation location and $K$ during the same injection test. It is apparent that the high correlation value covers much of the downstream area of the observation location and most of area left of the injection location. The result is consistent with that by [32]. A comparison between Fig. 5a and d indicates that a steady head measurement at a sampling location is correlated to $K$ field over a much larger area than a tracer moment measurement at the same location. Interestingly, Fig. 5a shows a very small region with absolute correlation value greater than 0.4 whereas Fig. 5d shows no region with absolute correlation higher than 0.4. These results explain the reason why hydraulic tomography can yield high resolution mapping of $K$ distribution over a large area and also why tracer data can complement the estimate of $K$ from hydraulic tests. These figures also indicate that, during a water/tracer injection test, the first moment is negatively related to $K$ and positively related to $\theta_w$ and $\theta_n$.

### 4. Discussion and conclusions

Using hydraulic/tracer tomography for characterizing DNAPL source zone is new but it faces a few challenges. One of them is

![Fig. 5. Cross correlations between the first moment perturbation and perturbations of: (a) $\ln K$; (b) $\ln \theta_w$; and (c) $\ln \theta_n$ and (d) cross correlation between perturbation of head and perturbation of $\ln K$. Where the filled red circle represents tracer/water injection location and open red circle is the tracer observation location, blue arrows denote the velocity field (not scaled to magnitude), and the black curves with arrows are the streamlines.](image)
the computational cost, which mainly arises from the usage of a large number of discrete concentration measurements during the estimation procedure, in which the solution to the hyperbolic advection–dispersion equation demands fine spatial and temporal discretizations. To reduce the computational cost associated with the new technology, the temporal moments of tracer BTCs are employed for parameter estimation in this study instead of point concentration measurements at some discrete times. Temporal moments are calculated using moment-generating equations. Solving these moment-generating equations is computationally more efficient than solving transient advection–dispersion equation because the temporal moments are time independent. Furthermore, the adjoint state equations have similar form as the corresponding forward equations, thus there is huge reduction in computational cost during the sensitivity calculation.

The results show that the estimates of $\theta_n$ from the first moments are not as good as those using discrete concentration data of the BTCs. This can be attributed to the fact that the first temporal moment of a BTC is an average value of the arrival times of concentration values of different parts of a BTC. The arrival time of each concentration value of a BTC reflects the travel path and heterogeneity that a volume of a tracer encountered during the injection and sampling points. Thus, the first moment only contains a portion of information in the BTC. Although higher moments potentially contain more information, the computational cost for using higher moments however will be significantly greater than the first moment since all moments have to be solved recursively. Nevertheless, estimates from the first moment reveal major high and low DNAPL content zones in the synthetic case, suggesting that the first temporal moments are useful for estimation of DNAPL source zones. The synthetic case also shows that the first moment data from one tracer data combined with hydraulic tomography can estimate DNAPL distributions with sufficient resolution while an increased number of tracer tests would increase the resolution of the DNAPL estimates but at greater costs.

Solving advection dominated moment-generating equations is prone to numerical oscillations. The oscillations occur during evaluation of both forward and adjoint state equations, which can lead to numerical instability during inversion. As the SUPG method is commonly used to reduce the oscillation, choosing an appropriate stabilization factor is critical. Our numerical simulations indicate that the one we used in conjunction with a solver from Diffpack works very well. With a Peclet number of approximately 10 in the synthetic cases, we do not experience convergence problems using the first temporal moment alone. However, small spurious oscillations still remain with the SUPG method and the oscillations will be expected using the SUPG method regardless choice of stabilization factors. Whether the oscillations could cause numerical instability for other cases has not been tested. Cirpka and Kitanidis [4] used a stabilization factor different than our choice for solving moment-generating equations, but they reported that inverse modeling using arrival time alone could lead to convergence problems. More elegant solving techniques that further reduce oscillation, such as adaptive mesh refinement, may be needed for more general applications.

The cross correlation analysis indicates that a tracer first moment measurement at one location contains information about parameters along a narrow strip between the injection location and the tracer measurement location while a steady head at the same location is correlated with hydraulic conductivity over a large area. Transient head measurements at later time of a pumping test exhibit similar cross correlation with hydraulic conductivity as steady state heads. One tracer test with a limited number of sampling locations therefore can only provide limited area coverage, making tracer moment measurements less efficient than hydraulic head measurements in estimating hydraulic conductivity. Furthermore, with all the simplifications and assumptions in this work for describing tracer movements through a DNAPL source zone, a tracer first moment measurement is still correlated not only to hydraulic conductivity but also to water content (conservative and partitioning tracers) and DNAPL content (partitioning tracer). A tracer measurement can also be affected by parameters such as diffusivity, partitioning coefficient and others, potentially further reducing the correlation between tracer first moment and hydraulic conductivity. Consequently, to yield a reasonable estimate of hydraulic conductivity using tracer measurements, one may consider conducting multiple tracer tests in a tomographic fashion, where each test provides different area coverage. However, conducting a tracer tomography survey in the field is potentially very costly.

Notice that cross correlation analysis is different from sensitivity analysis. Eqs. (27) and (28) show that calculating cross correlation between a state variable (i.e. tracer first moment) and a parameter (i.e. hydraulic conductivity) is the cross covariance normalized by the variance of the state variable and the variance of the parameter. The variance of the state variable in turn is affected by both the sensitivities of the state variables to all parameters and the covariances (or uncertainties) of all parameters (i.e., hydraulic conductivity, water content, and DNAPL content in this study). The cross correlation as demonstrated in this study is therefore an appropriate criterion for assessing the effectiveness of a measurement as well as different types of measurements on parameter estimation.

The current study assumes the source zones are located in confined aquifers while most real contaminations occur in unconfined aquifers. One of the major differences in applying hydraulic/tracer tomography to unconfined aquifers is on how to use head data collected from unconfined aquifers to estimate the spatial distribution of hydraulic parameters. The groundwater flow in unconfined aquifers is more complex due to the drainage or wetting process near the water table. Accurate representation of flow in unconfined aquifers may require saturated–unsaturated flow models in three dimensions [5, 24].

Finally, we must emphasize that this study is mathematically motivated to reduce the computational cost in interpreting hydraulic/tracer tomography data while the idea of tracer/tomography is still in the concept-developing stage. Many other issues related to the applicability of the new approach to real-world problems have yet to be addressed. These include: (1) the assumption that tracer only reacts with DNAPLs; (2) the assumption that the partitioning process is in equilibrium; (3) the assumption that DNAPLs are not mobilized by tracer tests; (4) memory requirement for large-scale problems. Nevertheless, our study demonstrates that using tracer temporal moments can dramatically reduce the computational cost in interpreting tracer tomography and can yield reasonable estimates of DNAPL content, making a step forward for applying hydraulic/tracer tomography to real-world problems.

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