Disparity of Macrodispersivity Estimated from Temporal and Spatial Moments Using Random Walk Particle Tracking in Heterogeneous Porous Formations

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Abstract: This paper presents advective and dispersive solute transport processes in naturally heterogeneous porous formations based on a random walk particle tracking method. Specifically, the disparity of macrodispersivities estimated from temporal moments of breakthrough curves at control planes and from spatial moments of particle cloud is investigated in non-uniform flow systems under the three different conditions of hydraulic conductivity distributions. For both temporal and spatial moment approaches, longitudinal macrodispersivity estimates in stratum porous formations are in considerable agreement with the results reported in the literature. The disparity of macrodispersivity attributes to the nature of both approaches to evaluate a travel distance of particle plume. While the second central temporal moments in stratum aquifer asymptotically approach a constant value regardless of the injection and pumping rates at active wells, the behavior of the second spatial moments depends upon the porous formations and pumping rates. Furthermore, spatial moment approach represents a potentially valuable tool in the assessment of transverse macrodispersion process in heterogeneous porous media.

Keywords: Random walk particle tracking; Macrodispersivity; Spatial moment; Temporal moment; Heterogeneity

1 Introduction

Solute transport in porous media is fundamental to many significant geoenvironmental problems. It is now well recognized that natural porous media exhibit spatial variability at the continuum scale and that this variability has a profound impact upon solute fate and transport at the larger field scale relevant to environmental and hydrogeological problems. Efforts to apply dispersive solute transport models under field conditions have demonstrated that field-observed dispersive coefficients can be several orders of magnitude larger than those indicated by laboratory tests with similar materials (e.g., Mackay *et al.*, 1986; LeBlanc *et al.*, 1991). The effect of spatial distribution of hydraulic conductivity on reactive and non-reactive solute transport is enhanced spreading, a phenomenon referred to as macrodispersion (Gelhar and Axness, 1983).

A theory of macrodispersion in aquifers based on a Lagrangian approach has been developed which links spatial and temporal moments and field measurable quantities such as hydraulic conductivity (e.g., Tompson, 1993; Lee *et al.*, 2007). Macrodispersivities are often estimated in the field by

monitoring breakthrough curves at a limited number of observation wells (Freyberg, 1986; Taylor and Howard, 1987). In a Lagrangian framework, temporal moments provide a measure of the non-Fickian behavior of transport in heterogeneous porous media and are therefore of concern. However, field tracer test, in general, based on breakthrough curves cannot infer transverse dispersivity. In the same manner as the temporal moments, a snapshot of a cloud of solute particles in time expresses the dispersive variation in the hydraulic conductivity field, leading to the estimates of spatial moments corresponding to the heterogeneity (Naff, 1990; Salamon *et al.*, 2006). Gelhar *et al.* (1992) illustrated the scale-dependence of longitudinal dispersivity, although no descriptions related to the difference between dispersivities derived from temporal and spatial moments were given. Relatively few investigators have considered the disparity of macrodispersivities derived from spatial and temporal moments in a field condition where large-scale variability is of importance as well as macrodispersivities under the differences between flow conditions associated with non-uniform flow paths taking the injection and extraction of groundwater into account.

The objective of this paper is to investigate the differences in the scale-dependence macrodispersivity estimates based on temporal and spatial moments in heterogeneous porous formations. The aquifer system investigated here relies on the hydrogeologic data in the southwest of the Netherlands and is modeled a homogeneous aquifer and two different hydraulic conductivity fields consisting of a stratified aquifer and a geostatistically-induced aquifer. Numerical tracer transport experiments using random walk particle tracking are conducted in these aquifers with injection and

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pumping at wells for estimating macrodispersivity in terms of spatial moments of particle positions and temporal moments of particle arrival times at control planes.

2 Advective-dispersive particle transport

2.1 Seepage flow

For the transport simulations, a saturated velocity field within a heterogeneous porous medium must initially be developed. The governing equation for hydraulic head distribution in a steady-state, three-dimensional aquifer system with recharge and discharge is usually written as (Bear, 1972)

$$\frac{\partial}{\partial x_i} \left(K(\mathbf{x}) \frac{\partial h}{\partial x_i} \right) = Q(\mathbf{x}) \quad (1)$$

where x_i are the Cartesian coordinates, $K(\mathbf{x})$ is the hydraulic conductivity at vector location $\mathbf{x}$, h is the hydraulic head at vector location $\mathbf{x}$, $Q(\mathbf{x})$ is the steady-state recharge and discharge at vector location $\mathbf{x}$. Summation over repeated indices is assumed. The local medium fluid velocity can be determined from

$$v_i(\mathbf{x}) = -\frac{K(\mathbf{x})}{n_p} \frac{\partial h(\mathbf{x})}{\partial x_i} \quad (2)$$

where $v_i(\mathbf{x})$ is the pore velocity at vector location $\mathbf{x}$ and n_p is the porosity.

2.2 Random walk particle tracking

Random walk particle tracking method is commonly used in the field of statistical physics to model and analyze processes involving diffusion. In the hydrologic community, the approach has been widely applied to simulate advective and dispersive (diffusive) mass transport problems in subsurface system (e.g., Tompson and Gelhar, 1990; Uffink, 1983; Inoue *et al.*, 2007).

Simulation of advective and dispersive mass transport may proceed by changing particle positions with time via an Itô-Taylor integral scheme, given as (Gardiner, 2004)

$$\mathbf{X}_p(t + \Delta t) = \mathbf{X}_p(t) + \mathbf{A}(\mathbf{X}_p, t)\Delta t + \mathbf{B}(\mathbf{X}_p, t)\mathbf{\Xi}(t)\sqrt{\Delta t} \quad (3)$$

where $\mathbf{X}_p(t)$ is the i -component of the particle location at time t , Δt is the time increment, and $\mathbf{\Xi}$ is a vector which contains three normally distributed random numbers with zero mean and unit variance. The vector $\mathbf{A}$ represents the deterministic drift determined by the fluid flow velocity and contains contributions from the dispersion tensor. The matrix $\mathbf{B}$ represents the direction displacement distance for the random process $\mathbf{\Xi}(t)\sqrt{\Delta t}$, which expresses Brownian motion.

Itô (1951) demonstrated that the particle density distribution $f(\mathbf{X}_p, t)$, which is defined as the probability of finding a particle within a given interval between $\mathbf{X}_p$ and $\mathbf{X}_p + d\mathbf{X}_p$ at a certain time t , fulfills in the limit of large particle numbers and infinitesimally small step size, the so-called Fokker-Planck equation. This equation describing the motion of the particle density distribution f is compared with the advection and dispersion equation and is given by the summation form (Uffink, 1990)

$$\frac{\partial f}{\partial t} = -\nabla \cdot (\mathbf{A}f) + \frac{1}{2} \nabla \nabla : (\mathbf{B} \cdot \mathbf{B}^T f) \quad (4)$$

In order to establish an analogy between the familiar advection and dispersion and the Fokker-Planck equations, the former can be expressed as follows (Zheng and Bennett, 1995):

$$\frac{\partial (n_p c)}{\partial t} = -\nabla \cdot (\mathbf{v} n_p c) + \nabla \cdot (n_p \mathbf{D} \cdot \nabla c) \quad (5)$$

where c is the dissolved concentration of solute, $\mathbf{v}$ is the pore velocity and $\mathbf{D}$ is a velocity-dependent dispersion tensor with the effective molecular diffusion given by as follows (Burnett and Frind, 1987a; Zheng and Bennett, 1995).

$$D_{11} = \alpha_L \frac{v_1^2}{v} + \alpha_{TH} \frac{v_2^2}{|\mathbf{v}|} + \alpha_{TV} \frac{v_3^2}{|\mathbf{v}|} + D_d \quad (6)$$

$$D_{22} = \alpha_{TH} \frac{v_1^2}{|\mathbf{v}|} + \alpha_L \frac{v_2^2}{|\mathbf{v}|} + \alpha_{TV} \frac{v_3^2}{|\mathbf{v}|} + D_d \quad (7)$$

$$D_{33} = \alpha_{TV} \frac{v_1^2 + v_2^2}{|\mathbf{v}|} + \alpha_L \frac{v_3^2}{|\mathbf{v}|} + D_d \quad (8)$$

$$D_{12} = D_{21} = (\alpha_L - \alpha_{TH}) \frac{v_1 v_2}{|\mathbf{v}|} + D_d \quad (9)$$

$$D_{13} = D_{31} = (\alpha_L - \alpha_{TV}) \frac{v_1 v_3}{|\mathbf{v}|} + D_d \quad (10)$$

$$D_{23} = D_{32} = (\alpha_L - \alpha_{TV}) \frac{v_2 v_3}{|\mathbf{v}|} + D_d \quad (11)$$

where α_L is the longitudinal dispersivity, α_{TH} and α_{TV} are horizontal and vertical transverse dispersivities, respectively, $|\mathbf{v}|$ is the magnitude of the velocity vector, and D_d is the effective diffusion coefficient. By using $\mathbf{A}$ and $\mathbf{B}$ as

$$\mathbf{A} = \mathbf{v} + \nabla \cdot \mathbf{D} + \frac{1}{n_p} \mathbf{D} \cdot \nabla n_p \quad (12)$$

$$2\mathbf{D} = \mathbf{B} \cdot \mathbf{B}^T \quad (13)$$

and noting that Eq.(4) and Eq.(5) become equivalent.

Substituting Eq.(12) and Eq.(13) into Eq. (3) yields the equation for the displacement of a particle in random walk (Uffink, 1990; Tompson, 1993; Salamon *et al.*, 2006).

$$\mathbf{X}_p(t + \Delta t) = \mathbf{X}_p(t) + \left(\mathbf{v} + \nabla \cdot \mathbf{D} + \frac{1}{n_p} \mathbf{D} \cdot \nabla n_p \right) \Delta t + \mathbf{B}(\mathbf{X}_p) \mathbf{\Xi}(t) \sqrt{\Delta t} \quad (14)$$

The displacement matrix $\mathbf{B}$ has the form (Burnett and Frind, 1987b; Lichtner *et al.*, 2002)

$$\mathbf{B} = \begin{pmatrix} \frac{v_1}{|\mathbf{v}|} \sqrt{2\alpha_L |\mathbf{v}|} & \frac{-v_1 v_3 \sqrt{2\alpha_{TV} |\mathbf{v}|}}{|\mathbf{v}| \Gamma} & \frac{-v_2 \sqrt{J}}{\Gamma} \\ \frac{v_2}{|\mathbf{v}|} \sqrt{2\alpha_L |\mathbf{v}|} & \frac{-v_2 v_3 \sqrt{2\alpha_{TV} |\mathbf{v}|}}{|\mathbf{v}| \Gamma} & \frac{v_1 \sqrt{J}}{\Gamma} \\ \frac{v_3}{|\mathbf{v}|} \sqrt{2\alpha_L |\mathbf{v}|} & \frac{\Gamma}{|\mathbf{v}|} \sqrt{2\alpha_{TV} |\mathbf{v}|} & 0 \end{pmatrix} \quad (15)$$

$$\Gamma = \sqrt{v_1^2 + v_2^2}, \quad J = 2 \left(\frac{\alpha_{TH} \Gamma^2 + \alpha_{TV} v_3^2}{|\mathbf{v}|} \right) \quad (16)$$

Equilibrium sorption using a linear isotherm can be incorporated by replacing the velocity $\mathbf{v}$ with a retarded velocity $\mathbf{v}/R$ and $\mathbf{D}$ with $\mathbf{D}/R$ in Eq. (14) where R is the retardation factor.

A second-order partial differential equation shown in Eq. (5) can be solved using an Eulerian approach by finite element method. However, in order to overcome the problem of artificial oscillation and numerical dispersion, the Peclet number and Courant number have to be sufficiently small. Random walk particle tracking offers free of these problems and can automatically satisfy global mass conservation due to the use of particles as discrete mass parcels.

2.3 Temporal moments

Temporal moments associated with observed breakthrough curves at several predefined control planes are computed using a Lagrangian framework. Monitoring the first passage time of particles passing through control planes allows for the estimation of temporal moments of breakthrough curves without having to evaluate the actual shape of the breakthrough curve. The n -th normalized absolute temporal moment can be calculated as the expected value of the arrival time of a particle at the control plane to the n -th power (Shapiro and Cvetković, 1988; Fernández-García *et al.*, 2005).

$$M_{n,T} = \frac{\sum_{k=1}^{NP_s} m_p^k (t_p^k(x_1))^n}{\sum_{k=1}^{NP_s} m_p^k} \quad (17)$$

where $M_{n,T}$ is the n -th normalized absolute temporal moment, x_1 is the mean flow direction coordinate, m_p^k is the mass assigned to the k -th particle, which is set constant for all particles due to the condition of no-decay, t_p^k is the first arrival passage time of the k -th particle, NP_s is the total number of particles arrived at the x_1 -control plane. The n -th normalized central temporal moment is calculated using relationship between central and absolute temporal moments,

$$\Lambda_{n,T} = \sum_{r=0}^n \binom{n}{r} M_{n-r,T}(x_1) (-M_{1,T}(x_1))^r \quad (18)$$

where $\Lambda_{n,T}$ is the n -th normalized central temporal moment.

Longitudinal macrodispersivity from temporal moments is calculated as (Goltz and Roberts, 1987)

$$A_{11}(\xi_P) = \frac{\xi_P}{2} \frac{\Lambda_{2,T}(\xi_P)}{(M_{1,T}(\xi_P))^2} \quad (19)$$

where A_{11} is the longitudinal macrodispersivity, ξ_P is the distance between the source and the x_1 -control plane. This is also applicable not only to conservative solutes but to reactive tracers. The advantage of particle tracking approach for computing the temporal moments as well as for identifying the longitudinal micro or macrodispersivity is that the absence of constructing the entire breakthrough curves and subsequent integration of concentrations over time (Valocchi, 1985; Inoue *et al.*, 2007).

2.4 Spatial moments

One of the important aims of transport theories is to predict the extent of dilution or mixing of a body of contaminated

water as it moves with the flow. A commonly used measure of dilution is the spatial moments of the concentration distribution. Spatial moments of aqueous concentrations distributed in space in a single realization are calculated from snapshots of particles at given times as follows (Kitanidis, 1988; Tompson, 1993).

$$m(t) = \sum_{k=1}^{NP_t} \frac{m_p^k}{R(X_p^k(t))} \quad (20)$$

$$X_{G,i} = \frac{1}{m(t)} \sum_{k=1}^{NP_t} \frac{m_p^k X_{p,i}^k(t)}{R(X_p^k(t))} \quad (21)$$

$$S_{ij}(t) = \frac{1}{m(t)} \sum_{k=1}^{NP_t} \frac{m_p^k X_{p,i}^k(t) X_{p,j}^k(t)}{R(X_p^k(t))} - X_{G,i}(t) X_{G,j}(t) \quad (22)$$

where $m(t)$ is the total liquid phase solute mass, m_p^k is the mass assigned to the k -th particle, $X_{G,i}$ is the first order spatial moment and means the center of mass of particles, S_{ij} is the liquid phase second spatial moments associated with the distribution of particles at a certain time, R is the retardation factor at the particle location, NP_t is the number of particles in the system at time t , and $X_{p,i}^k$ is the i -component of the k -th particle location.

Longitudinal and transverse macrodispersivities from spatial moments of the distributed particles are calculated as (Freyberg, 1986; Rajaram and Gelhar, 1993)

$$A_{11}(\xi_G(t)) = \frac{S_{11}}{2\xi_G(t)} \quad (23)$$

$$A_{22}(\xi_G(t)) = \frac{S_{22}}{2\xi_G(t)} \quad (24)$$

$$A_{33}(\xi_G(t)) = \frac{S_{33}}{2\xi_G(t)} \quad (25)$$

where A_{22} and A_{33} are the horizontal and vertical transverse macrodispersivity, respectively, $\xi_G(t)$ is the travel distance of the center of mass of the plume in the mean flow direction at time t . This is applicable to conservative as well as reactive solutes. Dispersivities from spatial moments obtained in this work are based on particle distributions at times when none of the particles has exited the domain.

3 Design of computational features

3.1 Study area and available data

This study site is located in the dunes near The Hague in the province of Zuid Holland, the Netherlands, as shown in Figure 1. Formations consisting of a Pleistocene deposits comprise the aquifer system having a total thickness of about 19 meters. At the top and bottom the aquifer is confined by thick clay layers, providing impervious layers.

A schematic view of the field of concern is illustrated in Figure 2. The hydraulic conductivity distribution in the aquifer at the experimental site has been studied by granular analysis of soil samples at positions denoted R_a and R_b in Figure 2 (Uffink, 1990). The resulting profiles of hydraulic

Figure 1: Location of the study area in the Netherlands.

Figure 2: Sketch of the flow domain and locations of the source and the control plane.

conductivity have the arithmetic mean of hydraulic conductivity of 1.07 m/day and the geometrical variance $\sigma_{\ln K}^2$ of 0.44 and are shown in Figure 3. Uffink (1990) assumed to be the stratification of this aquifer with the porosity of 0.34 in an entire domain and conducted random walk particle tracking in a two-dimensional cross-section of aquifer where an injection well Flip, an extraction well Flop and soil sample cores R_a and R_b are involved.

In this study, three types of hydraulic conductivity distribution are assumed to be isotropic and are modeled to investigate the effect of spatial variability of porous formations on the macrodispersion. One of the three such possible models is chosen to treat the hydraulic conductivity distribution as "Homogeneous aquifer". Additionally, like the Uffink model, the aquifer of interest is assumed to be the stratified aquifer in an entire three-dimensional field, referred to as "Stratum aquifer". On the other hand, soil sample cores at R_a and R_b provide vertical variations of hydraulic conductivity as shown in Figure 3. The experimental variogram of field data from Figure 3 is presented in Figure 4 by closed circles. The solid line indicates the fitted isotropic exponential model variograms calculated using Eq. (26) (Deutsch and Journel, 1992).

$$\gamma_K = n_K + c_K \left(1.0 - \exp \left(\frac{-h_K}{\lambda_K} \right) \right) \quad (26)$$

Figure 3: Hydraulic conductivity versus depth at two locations. Vertical distribution employed in this study is also depicted.

Figure 4: Experimental and fitted exponential variogram.

where γ_K is the variogram, c_K is the sill, h_K is the lag distance, n_K is the nugget effect and λ_K is the range. Parameter values of the fitted exponential model are set to $c_K = 0.35$, $\lambda_K = 2.0$, $n_K = 0$. As a few studies point out, the experimental variograms become noisy at larger lag distances owing to the small number of observations available (Welty and El-sner, 1997; Kobayashi *et al.*, 1998).

In order to interpolate values of hydraulic conductivity at unsampled locations in an entire domain, ordinary kriging is used (Journel, 1989; Deutsch and Journel, 1992), providing the third possible hydraulic conductivity distribution, referred to as "Geostatistics aquifer". In geostatistics aquifer, the variation of hydraulic conductivity is limited to a narrow range around soil sample points at R_a and R_b due to the variogram length. All aquifer models have the same mean value of hydraulic conductivity of 1.07 m/day but differ in porous formations. Figure 5 illustrates a profile of hydraulic conductivity distribution in a vertical section with $y = 80$ m for Stratum model and Geostatistical model.

3.2 Flow fields

A steady, saturated velocity field corresponding to each hydraulic conductivity field is developed by solving the system of Eq. (1) and Eq. (2) within the three-dimensional domain where prismatic grid spacing $\Delta x = 1$ m to 10 m, $\Delta y = 1$ m to 10 m and $\Delta z = 0.5$ m is adopted in each direction. The local head $h(x)$, i.e., the boundary condition of the first type, is specified on two sides faces intersecting the plane with $x = 0$

Figure 5: Hydraulic conductivity distributions: (left) Stratum model and (right) Geostatistical model. These graphs show the $x-z$ cross-section with $y = 80$ m.

and $x = 124$ m, while all other faces are treated as no-flow boundaries. This results in the establishment of a mean hydraulic gradient of 0.36 to reflect the pumping test reported in the literature (Uffink, 1990). As for solving the seepage equation, inflow is set to the positive in the finite element method.

Within each hydraulic conductivity distribution, three types of flow patterns are considered under the same boundary conditions for six faces. Injection and extraction wells are under the operation with a certain injection or pumping rate in the domain, and are located at Flip and Flop denoted in Figure 2, respectively. As an assumption, the total injection or pumping rate specified is divided evenly among the finite element nodes intersecting the well screen. As the base case, injection and extraction rates are set approximately to $Q(x) = 480 \text{ m}^3/\text{day}$ at Flip and $Q(x) = -480 \text{ m}^3/\text{day}$ at Flop, respectively, in order to compare the difference of macrodispersivity outcomes between estimates of this study and data reported in the literature (Uffink, 1990). Therefore, flow fields exhibit the “doublet” or convergent flow (Huyakorn *et al.*, 1986) in the vicinity of an injection and pumping well pair, despite of the porous formations. Moreover, two different operation conditions of both wells at one-half and one-tenth the total rate are considered to investigate the effect of the total rate of pumping or injection on the macrodispersivity estimates. All other specifications including the boundary conditions remain unchanged from the base case.

3.3 Particle tracking implementation

To implement any particle tracking algorithm, a method is needed for computing the velocity vector at all locations in the model domain in order to calculate the deterministic portion of the particle transport vector, given by $\mathbf{A}$ in Eq. (12). In addition, application of the random walk particle tracking requires knowledge of the velocity gradient at particle location. However, when a numerical flow model such as finite element model is used to solve for the head distribution, groundwater velocity is known only at certain locations. Therefore, in this study, velocity interpolation technique (Cordes and Kinzelbach, 1992; Zheng and Bennett, 1995) is employed to compute the components of flow velocity associated with an arbitrary location of a particle. While a finite element model

Table 1: Parameters used in this study.

Longitudinal dispersivity (m)	0.01
Horizontal transverse dispersivity (m)	0.001
Vertical transverse dispersivity (m)	0.001
Effective diffusion coefficient (m^2/day)	0
Retardation factor	1
Porosity	0.34
Number of particles	2×10^5
Time increment (day)	1
Injection rate at well (m^3/day)	48, 240, 480
Pumping rate at well (m^3/day)	48, 240, 480

is the computational framework on which particle tracking methods are based, random walk particle tracking method stated above is performed under the prescribed flow conditions.

As an initial pulse input, the number of particles used to represent the solute is 2×10^5 with a constant mass of $76800 \text{ g}/2 \times 10^5$ per one particle in order to reflect the mean concentration of pumped water at this site (Uffink, 1990). All particles are applied uniformly distributed at random within the injection well, Flip, as shown in Figure 2. Additionally, longitudinal dispersivity and horizontal and vertical transverse dispersivities are chosen to be $\alpha_L = 0.01 \text{ m}$, $\alpha_{TH} = 0.001 \text{ m}$ and $\alpha_{TV} = 0.001 \text{ m}$, respectively, which are based on the data reported in the literature (Uffink, 1990). When a particle arrives at a withdrawal well Flop, that particle is regarded to flow into the well and is removed from the flow domain. To simplify the problem, a particle is assumed to be conservative and not to have the adsorptive nature, leading to 1 of retardation factor. Parameters used in this analysis is listed in Table 1.

Furthermore, application of a random walk particle tracking requires that some computational approximations be established. The time step used to move a particle through space is required to be small enough because use of too large a time step may lead to overshoot errors in the displacement expressed in Eq. (14) (Tompson *et al.*, 1995). Thus, whereas all particles move from one time level to another with an identical time step value and are allowed to transfer and disperse until a maximum duration of 1000 days, step doubling procedure (Zheng *et al.*, 1991) is used to assure the accuracy of particle tracking linked with the fourth-order Runge-Kutta method.

Swarm of particles is advected and dispersed according to the velocity distribution toward a (y,z) -plane located at some distance from the injection location. This study focuses on the passage of particles through a (y,z) -plane, referred to as the control plane, through which the solute mass flux is to be predicted or measured. For a quantitative description of the transport and macrodispersion of particles, the breakthrough of the solute plume across the entire control plane is assessed. Several control planes are located from $x = 28 \text{ m}$ to $x = 99 \text{ m}$ in the x -direction to evaluate the particle behavior passing through these control planes, as seen in Figure 2. Break-

Figure 6: Comparison of longitudinal macrodispersivity estimated from temporal moments of breakthrough curves and spatial moments of tracer plume under the pumping rates of 480 m³/day.

Figure 7: Effect of the degree of extraction rate at the pumping well on the longitudinal macrodispersivity under the pumping rates of 240 m³/day.

Figure 8: Effect of the degree of extraction rate at the pumping well on the longitudinal macrodispersivity under the pumping rates of 48 m³/day.

through curves obtained at each control plane allow to estimate the longitudinal macrodispersivity based on Eq. (19).

On the other hand, the overall development of a solute plume is able to be measured in terms of spatial moments of its mass distribution using Eq. (20) through Eq. (22). Contrary to the macrodispersivity estimated from temporal moments, spatial moment analysis is a useful tool to identify not only the longitudinal macrodispersivity but the transverse macrodispersivities using Eq. (23) and Eqs. (24) and (25), respectively. This point is one advantage of a particle tracking approach to effectively achieve the estimate of macrodispersivity in a heterogeneous flow field instead of the use of conventional finite element or finite difference techniques.

4 Results and discussion

4.1 Influence of the hydraulic conductivity distribution

Longitudinal macrodispersivity estimates from temporal and spatial moments normalized by the local or microdispersivity as a function of the travel distance along x -direction from the injection well is illustrated in Figure 6 under the absolute pumping/injection condition of 480 m³/day. Reported estimates (Uffink, 1990) at three points are also plotted in Figure 6. Although all simulations run for a period of time long enough to construct a relatively complete downstream breakthrough curve, spreading and displacement characteristics in terms of spatial moments of the particle distribution may be limited to a certain time period because all particles are required to exist within the domain to ensure the accuracy of statistical moments. According to the porous formation and the activity of well, an arrival time of particle to the pumping well varies significantly, leading to the difference of macrodispersivity evolution estimated from spatial moments.

As shown in Figure 6, macrodispersivity estimates range from 10² to 10³ and are almost identical to the results obtained by Uffink, despite of the hydraulic conductivity distribution. Whereas both temporal and spatial moments provide the low degree of macrodispersivity increase as well as the results of Uffink in stratum aquifer, while the change

of macrodispersivities both for geostatistics and homogeneous aquifers remains approximately constant. Generally, whereas typical values of local dispersivity in laboratory experiments range between 0.001 and 0.1 m (e.g., Xu and Eckstein, 1997; Guedes de Carvalho and Delgado, 2003; Inoue *et al.*, 2006), values of macroscopic dispersivity are two to four orders of magnitude larger (e.g., Neuman, 1990; Gelhar *et al.*, 1992). This disparity arises because dispersion at the field scale is caused primarily by macroscopic heterogeneities, rather than by pore-scale heterogeneity. Field-scale dispersivity increases with distance and with time and is influenced to the stratification of an aquifer (Gelhar and Axness, 1983; Uffink, 1983). Consequently, Figure 6 demonstrates that evolution of macrodispersivities from both temporal and spatial moments in stratum aquifer is appropriate.

In order to investigate the effect of the degree of pumping rate at withdrawal well, macrodispersivity estimates for all hydraulic conductivity distributions as a function of the travel distance under the pumping rates of 240 m³/day and 48 m³/day are plotted in Figures 7 and 8, respectively. When a pulse of solute mass introduces into a flow field behavior of macrodispersivity is erratic with time, particularly very close

Figure 9: Disparity between macrodispersivities estimated from temporal and spatial moments: (top) plan view of the particle cloud illustrating the location of the control plane and (bottom) vertical view of the particle cloud with the center of mass.

to a source (Molz *et al.*, 1983; Tompson and Gelhar, 1990). As the plume continues to move, it will expand and gradually be characterized after a certain displacement occurs. In stratum aquifer, macrodispersivities from temporal moments approach an asymptotic value, while macrodispersivities from spatial moments increase with the increase of travel distance and are in good agreement with the results of Uffink (1990). It can be observed that stratum aquifer exhibits less sensitive to flow paths when computing the macrodispersivity. Thus, in layered porous formations macrodispersivity can be estimated using either temporal or spatial moments despite of the pumping rate of the injection and extraction wells.

On the other hand, variation of macrodispersivities in geostatistics and homogeneous aquifers is substantially sensitive to the change of pumping rates. As for the case with $48 \text{ m}^3/\text{day}$ of pumping rate, values of macrodispersivities are approximately 1.0, except for a range within a relatively short travel distance, indicating that macrodispersivity is identical to microdispersivity. In geostatistics aquifer of interest, behavior of particles appears to be similar to that in homogeneous aquifer due to the less heterogeneity of aquifer with 0.44 of geometric variance as shown in Figure 5. Therefore, the structure of hydraulic conductivity distribution in an entire domain in geostatistics aquifer may be regarded as a homogeneous formation, although the hydraulic conductivity varies partially in the vicinity of sampling positions R_a and R_b . Moreover, as pointed out by Goltz and Roberts (1987) and Inoue *et al.* (2007), both temporal and spatial moments

Figure 10: Variation of cumulative mass of particles at control planes in geostatistics aquifer.

Figure 11: Variation of cumulative mass of particles at control planes in stratum aquifer.

are capable of providing the microdispersivity in homogeneous porous formation under a uniform flow condition. The pumping rate of $48 \text{ m}^3/\text{day}$ is approximately 3% of the rate of flow in volume of water of the domain and appears to be relatively small. Hence, when a convergent, or non-uniform flow may be treated as a uniform flow in geostatistics and homogeneous aquifers of concern under the low pumping rate condition such as $48 \text{ m}^3/\text{day}$ in this study, temporal and spatial moments are capable of assessing the microdispersivity, instead of the macrodispersivity.

4.2 Disparity of macrodispersivity

As seen in Figures 6 through 8, in stratum aquifer, macrodispersivities estimated from spatial versus temporal moments exhibit different scale-dependent behavior, while the influence of pumping rate on the asymptotic values estimated from spatial and temporal moments is not significant. As pointed out by Dagan (1984) and Naff (1990), the asymptotic constant values are expected to be almost the same for both moments of macrodispersivities as travel distance increases. However, there are differences in the distance-scale at which the macrodispersivities achieve a constant value in aquifer of concern. It is evident from Figures 6 through 8 that the distance-scale at which macrodispersivities from temporal moments of breakthrough curves at a control plane ap-

Figure 12: Variation of the first absolute and second central temporal moments as a function of location of control planes in geostatistics aquifer.

Figure 13: Variation of the first absolute and second central temporal moments as a function of location of control planes in stratum aquifer.

proach an asymptotic value is smaller than the corresponding distance-scale for macrodispersivities estimated from spatial moments of particle cloud. The degree of this disparity between macrodispersivities estimated from temporal and spatial moments is in common regardless of a prescribed pumping rate.

Figure 9 illustrates the particle cloud of a tracer in plan and vertical views as one example to explain the behavior for stratum aquifer under the pumping condition of 240 m³/day. To simplify a snapshot of particle distribution, a swarm of 10⁴ particles 30 days after injection and its centroid in *x*, *y* and *z* coordinates are plotted. A breakthrough curve assessed at a control plane described in Figure 9 provides almost the same macrodispersivity that obtained from the second spatial moment corresponding to a specific plume centroid location, also shown in the figure. All particles applied at the source, i.e., Flip in this study, are required to pass through a specific control plane to express a complete breakthrough curve and to lead macrodispersivity using temporal moments. On the contrary, application of spatial moment method is restricted since all particles emerged at the injection point are necessary to identify the estimates. Whereas spatial moment

method is capable of leading a value as macrodispersivity at a corresponding travel time as long as all particles exist in the aquifer, larger times are necessary to assess an appropriate macrodispersivity based on temporal moment method. If at larger times as mentioned above, both temporal and spatial moments could provide the same asymptotic macrodispersivity despite of the flow condition, as reported by Dagan (1986) and Naff (1990). Hence, in flow fields in this study, due to a restriction of travel distance of particles, macrodispersivity estimates from temporal moments become larger than those from spatial moments.

4.3 Characteristics of temporal and spatial moments

The relative cumulations of mass breakthrough curves divided by the total mass of application at the source in geostatistics aquifer are shown in Figure 10 as for representative two control planes for all pumping rate conditions. It can be observed that the number of particles passing through a control plane varies abruptly, especially for the case with the smallest pumping rate. A time series mass variation indicates that breakthrough curves exhibit a similar Gaussian shape regardless of the location of a control plane, although a slight tailing of breakthrough curves can be seen for the largest pumping rate. This point attributes to the similarity of geostatistics aquifer of interest to the homogeneous aquifer where the evolution of cumulative mass of particles shows a Gaussian distribution (Welty and Gelhar, 1994; Inoue *et al.*, 2007).

In contrast to the results depicted in Figure 10, variation of cumulative mass of particles at two control planes in stratum aquifer is shown in Figure 11. It is apparent from the plot that the number of particles arriving at a control plane gradually increases and varies in a stepwise fashion after approximately half the number of particles pass through the control planes. This plume behavior reflects the hydraulic conductivity distribution on which the evolution of plume is considerably dependent.

As stated above, alike from Eulerian approach such as finite element or finite difference method, construction of the entire breakthrough curves and subsequent integration of mass variation with respect to time are not required to evaluate temporal moments in particle tracking approach. Temporal moments associated with each control plane are calculated and utilized to determine the longitudinal macrodispersivity (Valocchi, 1985; Goltz and Roberts, 1988). To quantify the values of temporal moments, the first absolute temporal moments $M_{1,T}$ in Eq. (17) and the second central temporal moments $\Lambda_{2,T}$ in Eq. (18) in geostatistics and stratum aquifers are plotted in Figures 12 and 13, respectively.

The most noticeable quantitative difference is relevant to the variation of the second central temporal moments denoted by closed symbols in these figures. While the second central temporal moments $\Lambda_{2,T}$ in geostatistics aquifer vary corresponding to the pumping rate as well as the location of control plane, it is interesting to note that stratum aquifer provides an asymptotic value of the second central temporal moments regardless of the pumping rate, except for a location relatively closer to the source. It is inferred that as for

Figure 14: Evolution of the second spatial moments as a function of the first spatial moments under different pumping conditions: (a) S_{11} , (b) S_{22} and (c) S_{33} .

stratum aquifer, the effect of hydraulic conductivity distribution on plume evolution is much more pronounced compared to the effect of pumping rate because plume behavior in aquifer depends highly not only on the hydraulic conductivity of a layer at which a particle is located but also on the vertical mixing of particles between layers (Uffink, 1983; Hoteit *et al.*, 2002). Hence, eventually, the marked difference of asymptotic values between geostatistics and stratum aquifers can be observed in Figures 6 and 7.

In order to characterize the evolution of particle swarm in each direction, the principal components of the tracer plume spreading variance tensor S_{ii} are plotted as a function of the first spatial moment $X_{G,1}$ in x -coordinate for all pumping rate conditions in both geostatistics and stratum aquifers in Figure 14. These components can be used to gauge the size of a particle cloud in a given direction and determine the rate

Figure 15: Comparison of horizontal and vertical transverse macrodispersivities estimated from spatial moments under the pumping rate of 480 m³/day in geostatistics and stratum aquifers.

of growth as a function of mean displacement. They may be used to assess the nature of macrodispersion, and in particular, to determine whether constant macrodispersivity components can be identified and measured.

The predominant axis of spreading and the nonlinearity of the growth occur in the longitudinal direction for stratum aquifer (Figure 14(a)). Transverse spread variance perpendicular to the longitudinal axis shown in Figure 14(b) has its own asymptotic value, while that in the vertical direction (Figure 14(c)) exhibits still another behavior. This means that velocity distribution and porous formation in space contribute to the elongation of particles in each direction. As a whole, each set of curves indicates a marked degree of variability corresponding to the flow condition. Increase or decrease of the second order spatial moments may reflect the stretching and squeezing each particle cloud undergoes during displacement. These different growth may be due to a developmental process (Dagan, 1984) associated with generated plume expansion, variabilities associated with the plume moving through different hydrogeologic layers, or a combination of both (Tompson *et al.*, 1995).

The transverse macrodispersivity components A_{22} and A_{33} estimated from spatial moments using Eq. (24) and Eq. (25) and normalized by local dispersivities of α_{TH} and α_{TV} , respectively, are plotted as a function of displacement distance in Figure 15. Since the variation of the transverse macrodispersivities does not depend upon the pumping rate, estimates under the condition of pumping rate of 480 m³/day are shown as a representative outcome. Both horizontal and vertical transverse dispersivities decrease with the increase of travel distance of particle cloud and eventually reach more than three and two order of magnitude larger than local horizontal and vertical transverse dispersivities, respectively. Dagan (1986) and Rajaram and Gelhar (1993) suggest the asymptotic value of the transverse macrodispersivities is zero under uniform flow condition. It is interesting that under non-uniform flow condition nonzero values of A_{22} and A_{33} are obtained as far as travel distance of particles is bounded and is in the range of approximately 100 m. Nevertheless,

contrary to the use of temporal moments, it is emphasized that spatial moments allow to estimate not only the longitudinal macrodispersivity but the transverse macrodispersivity. In the assessment of the fate and transport of contaminants in a field, as hydraulic properties of subsurface materials cannot be fully characterized in a deterministic way, stochastic methods and modeling of the data must be invoked. Stochastic analysis such as random walk particle tracking provides the reliable macrodispersion relevant to the variability and the spatial structure associated with hydraulic properties of the heterogeneous medium under different flow conditions.

5 Conclusions

Disparity of macrodispersivities estimated from temporal moments of mass breakthrough curves and spatial moments of particle plumes has been investigated through random walk particle tracking approach in homogeneous and heterogeneous porous formations, which reflect the soil samples in the field in the southwest of the Netherlands. The variation of macrodispersivity under non-uniform, or convergent flow conditions linked with the activity of injection and withdrawal wells has been focused on.

For stratum model, as the travel distance increases, longitudinal macrodispersivities from temporal and spatial moments increase and are in good agreement with the results of Uffink (1990). Dispersive processes occurring in the aquifer due to heterogeneity are captured differently in breakthrough curves at control planes than in snapshots of particle cloud. When macrodispersivities are assessed based on observed breakthrough curves, all particle generated in the aquifer are required to pass through a given control plane, leading to the disparity of macrodispersivities between temporal and spatial moments.

On the other hand, temporal and spatial moments in homogeneous and geostatistics aquifers of concern based on the soil samples in the field provide almost the same variation of macrodispersivities due to less heterogeneity of the aquifer. Additionally, it is revealed that stratum aquifer is less sensitive to flow paths when computing the macrodispersivity, although the values of macrodispersivity in homogeneous and geostatistics aquifers depend highly on the pumping rates not on the structure of porous formations.

The variation of the temporal moments as well as the spatial moments has been evaluated as quantitative measures of moving particles in the system. The evolution of particle cloud is substantially affected by the hydraulic conductivity distribution. It is worth noting that the second central temporal moments in stratum aquifer approach an asymptotic value despite of the pumping rate at larger travel distance, while the degree of growth with regard to the second spatial moments varies considerably under a corresponding condition, reflecting the effect of velocity distribution and porous formation in space on the elongation of particles in each direction. Moreover, the spatial moments of particle plumes serve as a tool for estimating not only the longitudinal macrodispersivity but the transverse dispersivity. This point indicates the possibility of their application to field tests in the context of predicting the behavior of large contaminant plumes.

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