

Mixing processes in highly heterogeneous formations

FRANCESCA BOSO & ALBERTO BELLIN

Dipartimento di Ingegneria Civile e Ambientale, Università degli Studi di Trento, Italy
francesca.boso@ing.unitn.it

Abstract We numerically explored the interplay between formation heterogeneity and local dispersion on the transport of a passive tracer in highly heterogeneous formations. In order to minimize the negative impact of numerical diffusion, we used the Smoothed Particle Hydrodynamics scheme recently proposed by Herrera *et al.* (2009). Heterogeneity enhances mixing, as measured by the dilution index, with a rate of increase with the logtransmissivity variance that attenuates passing from moderately to highly heterogeneous formations. In addition, the sample frequency distribution of the solute concentration is well represented by a Beta model at both low and high variances. The Beta model requires the knowledge of the spatial mean and variance of the solute concentration as the only parameters to be determined.

Key words mixing; highly heterogeneous formations; concentration distribution

INTRODUCTION

The distribution of solute concentration within a plume transported in a heterogeneous saturated formation is known to be very disordered and dependent on the complex and nonlinear interplay between local dispersion and spatially variable advection. Given this complexity, most of the early studies dealt with the ensemble mean concentration under the assumption that hydraulic property variations can be described as Stationary Random Space Functions (RSFs). However, the ensemble mean concentration provides a very poor representation of the real distribution of the solute concentration, particularly at the fringes of the plume. The situation is even worse for multispecies solute transport because the reaction between species in solution is typically nonlinear and sensitive to local concentration. Therefore, as clearly evidenced by Kapoor *et al.* (1997) using in the reaction terms the ensemble mean concentrations instead of the unknown local concentration leads to large errors in the global transformation rate. A large body of literature showed that mixing is the dominant mechanism in the transformation of aqueous species transported in heterogeneous formations and global measures have been proposed, such as the dilution index (Kitanidis, 1994) and its generalization (Rolle *et al.*, 2009) and the segregation intensity (Kapoor *et al.*, 1997), to describe the disorder of the concentration field and how far the local concentration is expected to be from the ensemble mean. These global quantities are important to assess how representative of solute transformation is the reaction rate computed with the ensemble mean concentration of the reacting species, but are of little use in applications. In this respect, we observe that much less work has been done to compute the spatial distribution of the concentration within the plume, despite the fact that the frequency distribution of these concentrations is itself a global measure of mixing and is usable in risk assessment. The frequency distribution can be used, for example, to obtain the probability of exceeding a target concentration at any position within the plume. This is incomplete, but important, information for risk assessment, which is more reliable than performing risk assessment with the ensemble mean concentration as is currently done in most applications.

In this work we numerically analyse the frequency distribution of the solute concentration within an ergodic plume, showing that it can be obtained as a function of spatial moments of the solute concentrations at both weakly and highly heterogeneous formations.

SIMULATION SET-UP AND NUMERICAL SCHEMES FOR FLOW AND TRANSPORT

We simulated flow and transport of a passive tracer and two aqueous species reacting upon mixing in a 2D heterogeneous formation with a Gaussian distribution of the hydraulic log-transmissivity $Y = \ln T$, where T is the hydraulic transmissivity. We adopted the typical Gaussian model of spatial

variability, which assumes the Y as a Random Space Function (RSF) with mean m_Y and variance σ_Y^2 , both constant, and the exponential isotropic covariance function:

$$C_Y(r_1, r_2) = \sigma_Y^2 \exp\left[-\frac{\sqrt{r_1^2 + r_2^2}}{I_Y}\right] \quad (1)$$

where I_Y is the log-transmissivity integral scale. Simulations are performed with σ_Y^2 varying between 0.2 and 10, i.e. from weakly to strongly heterogeneous formations in a squared flow domain with side equal to $L = 100 I_Y$.

A single realization of the log-transmissivity field is generated on a computational squared grid with cell size equal to $\Delta = 1/12 I_Y$ by using Hydro_Gen, a RSF generator developed by Bellin & Rubin (1996) as an evolution of the classical Sequential Gaussian Simulator (SGS).

The governing equation for the hydraulic head h is as follows:

$$\frac{\partial}{\partial x_1} \left[T(x_1, x_2) \frac{\partial h(x_1, x_2)}{\partial x_1} \right] + \frac{\partial}{\partial x_2} \left[T(x_1, x_2) \frac{\partial h(x_1, x_2)}{\partial x_2} \right] = 0 \quad (2)$$

Boundary conditions are of imposed hydraulic head at the upstream and downstream sides of the computational domain and no flow conditions at the remaining two sides, parallel to the mean flow direction. In addition, the vertically averaged velocity field $\mathbf{v} = (v_1, v_2)$ is given by the Darcy's law:

$$v_i(x_1, x_2) = \frac{T(x_1, x_2)}{n b} \frac{\partial h(x_1, x_2)}{\partial x_i}, \quad i = 1, 2 \quad (3)$$

where n is the formation's porosity, assumed constant throughout the computational domain, and b is the aquifer's thickness.

We solved equations (2) and (3) by using MODFLOW (Harbaugh *et al.*, 2000), which implements a finite volume scheme with equal squared cells of side $\Delta = I_Y/12$. The velocities within the computational cells are interpolated linearly by using the edge values in order to obtain a locally conservative continuous velocity field (La Bolle *et al.*, 1996).

The resulting velocity field is used to simulate transport of a solute instantaneously injected into a large initial volume $0.25 I_Y$ long and $80 I_Y$ large (according to the adopted conceptual model the injection is uniform over the aquifer's thickness) centred at $\bar{a} = (5 I_Y, 50 I_Y)$. Finally, local pore-scale dispersion D is set such as to obtain a Peclet number of $Pe = U I_Y / D = 1000$ in both longitudinal and transverse directions. In the definition of the Peclet number we used the effective mean velocity $U = Q/(nL)$, where Q is the total flux entering through the upstream boundary of length L .

Transport of a non reactive tracer is governed by the following mass balance equation:

$$\frac{\partial u}{\partial t} + \mathbf{v} \cdot \nabla u = \nabla \cdot (D \nabla u) \quad (4)$$

where $u = C/C_0$ is the dimensionless solute concentration with respect to the initial concentration C_0 of the solute injected instantaneously at the initial time $t = 0$. The solution of equation (4), with a suitable initial condition, can be substituted in an algebraic speciation equation to obtain the concentrations of two aqueous species, A and B, reacting upon mixing to produce a precipitate C according to the bimolecular reaction $A + B \rightarrow C \downarrow$ (see e.g. Valocchi *et al.*, 1981; Rubin, 1983). As well documented in literature (see e.g. Kapoor & Kitanidis, 1997) bimolecular reactions are strongly dependent on mixing, with the reaction that does not occur when local dispersion is turned off. Therefore, the importance of studying how mixing depends on the interplay between non-uniform advection and local dispersion goes beyond the interest for transport of a passive tracer to involve cases in which two aqueous species react upon mixing.

Given the importance of correctly reproducing mixing, in the simulations it is crucial to adopt numerical schemes not affected by artificial (numerical) diffusion. Among the different schemes available we chose to work with the Smoothed Particle Hydrodynamics algorithm (Monaghan, 2005) applied to equation (4) as proposed by Cleary & Monaghan (1999) and Tartakovsky *et al.* (2007, 2008). In particular, Herrera *et al.* (2009) applied SPH to the solution of equation (4) at the

Darcy scale. According to this meshless scheme concentration is computed by means of kernel integral interpolation on a moving set of nodes, which represent fluid particles. The inflow along the upgradient boundary is modelled through the injection of particles proportionally to the local velocity. The advective component of equation (4) is obtained by particle tracking, which is inherently free of numerical diffusion, whereas local dispersion is added through an approximation of the local diffusive fluxes among particles with different concentration.

RESULTS AND DISCUSSION

In the following we present a few results of our numerical simulations with the aim of investigating how the interplay between spatially-variable advection and local pore-scale dispersion influences the spatial distribution of the solute concentration within an ergodic plume. Results are sensitive to how the solute is introduced, in particular at high heterogeneity. In the present work we consider uniform injection within the initial volume because flux proportional injection, although resembling more closely most field situations, results in an initial concentration variance that varies (increases) with σ_Y^2 , thereby hindering the effect of heterogeneity on the concentration distribution and other global measures of dilution, such as the dilution index.

Dilution index

A widely used global measure of dilution is the dilution index E :

$$E(t) = \exp \left[- \int_{\Omega} p(x,t) \ln(p(x,t)) dx \right] \quad (5)$$

where $p(x,t) = c(x,t) / \int_{\Omega} c(x,t) dx$ is the ratio between the point concentration and the total mass of injected solute. Kitanidis (1994) showed that E is proportional to the volume of the plume, thereby providing a reliable global measure of dilution in the case of instantaneous injection when the total mass of solute is constant. It can be shown that E is sensitive to both local dispersion and the deformation of the plume with the latter which depends on how advection changes in space. Therefore, for a given local dispersion it is expected that the stronger the heterogeneity of the velocity field, the larger the dilution because of larger concentration gradients distributed through the plume. Figure 1(a) shows the evolution of E for a passive solute and for several σ_Y^2 values. For all σ_Y^2 considered in the present work E increases less than linearly at early times, and linearly at later times. This late time behaviour is in agreement with asymptotic analysis of Kitanidis (1994), which concluded that at large times $E \propto t^{N/2}$, where N is the space dimensionality. The nonlinear behaviour at early times is much more pronounced at larger σ_Y^2 values. On the other hand, for a given dimensionless time, E increases with σ_Y^2 , but at a rate that attenuates progressively with σ_Y^2 . Small to negligible differences are observed for $\sigma_Y^2 \geq 4$, suggesting an upper limit to the increase of the concentration gradients created by the plume's distortion. At weak to moderate heterogeneity an increase of σ_Y^2 results in larger concentration gradients but does not change significantly the shape of the plume, except that it becomes more elongated. In this situation the plume develops thin fingers along interconnected paths with relatively high velocity, resulting in spatially well distributed high concentration gradients across these fingers. In contrast, in highly heterogeneous formations the plume tends to split into separate portions, and the transverse size is larger than the fingers observed at lower heterogeneity. These portions tend to remain compact within channels, with relatively high velocity showing large dilution at the fringes, where the concentration gradients are large, but with small or almost no dilution inside, where the concentration varies much less than in the corresponding weakly heterogeneous formations. This polarizing of mixing around two extremes of small to negligible mixing and very large mixing has the final effect of reducing the increase of disorder, as measured by E . In our simulations this kinematic limiting factor of the growth of E is evident for $\sigma_Y^2 \geq 4$. The rather regular distribution of the velocity field, that characterizes the weakly heterogeneous formations, leads to a small dilution index, which shows a moderate linear increase

with time, also at the early times. In addition, the slope of the linear portion of the curve increases with σ_Y^2 with the tendency of reaching a constant asymptotic value for $\sigma_Y^2 \geq 4$, and this is more evidence that disorder grows less with σ_Y^2 for highly heterogeneous formations.

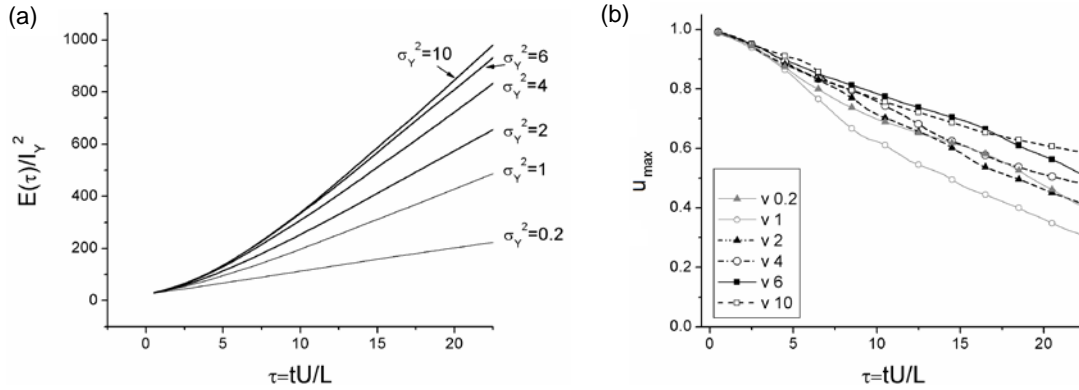


Fig. 1 Dilution index (a) and maximum solute concentration (b) versus the dimensionless travel time for σ_Y^2 ranging from 0.2 to 10.

Maximum concentration

In Fig. 1(b) we show the time evolution of the maximum concentration of a passive solute, which is also a quantity of interest in risk assessment, and an index of dilution (Fiori, 2001). The inherently local nature of the maximum concentration makes it more sensitive to local dynamics than global indexes such as the dilution index or the scalar dissipation rate.

At the early times the maximum concentration reduces at a rate that is independent on σ_Y^2 , but starting from $\tau = 5$ the effect of the formation's heterogeneity starts to manifest its effect. For small to moderate heterogeneity, i.e. for $\sigma_Y^2 \leq 1$ the rate of reduction of u_{max} increases with σ_Y^2 . This is an expected result since the plume dilutes faster in more heterogeneous formations, as an effect of plume deformation induced by spatially variable advection. However, we also observe that for $\sigma_Y^2 \geq 1$ the maximum concentration reduces less with a reversed general trend showing smaller reduction with higher σ_Y^2 . This result is consistent with what we observed for the dilution index and may reflect the fact that in highly heterogeneous formations portions of the plume may remain entrapped in low conductive zone where concentration gradients are smaller and therefore mixing is locally smaller. This leads to an attenuate reduction of the maximum concentration with results affected by the local variability of low hydraulic transmissivity zones, which can also be low in highly heterogeneous formations. This suggests that the maximum concentration can be affected by the model of spatial variability and in particular by the distribution of low and high transmissivity zones and their internal variability.

Spatial variability of concentration of a non reactive tracer

The dilution index is a global indicator of how the concentration of a passive tracer is expected to reduce with time as an effect of mixing, which is controlled by the interplay between local dispersion and the plume's deformation, the latter controlled by advection. However, this quantity is of difficult use in practical applications, where the typical question asked is if somewhere within a plume, or at a given target, the concentration can exceed a given threshold. The Cumulative Frequency Distribution (CFD) $F(C \leq c)$ can be used for this purpose. It represents the probability that the concentration is below a given value c in any point within the plume, or in other words, the fraction of the plume's volume with concentration below c . The complement to 1 of F represents the fraction of the plume with concentration higher than a give threshold, which is a valuable piece of information, although not exhaustive, in applications. Notice that $F(C \leq c)$ is

inherently different from the Cumulative probability Distribution Function (CDF) of point concentration, which is computed by statically averaging over independent realizations of the transmissivity field. The CDF shows dramatic space variability and reflects mainly uncertainty in the point concentration, which is very high except at exceedingly high times, when local dispersion overwhelms macrodispersion and the plume becomes Gaussian (Fiori & Dagan, 2000).

In a recently published paper Bellin & Tonina (2007) observed that in weakly to moderate heterogeneous formations the frequency (space) distribution of the concentration of a passive solute is well represented by a Beta distribution with the parameters depending on the spatial mean and variance of the solute concentration. In this work we numerically verify that the Beta model of F holds also for large and very large heterogeneous formations

According to the Beta model the concentration *pdf* assumes the following expression:

$$f_z(u) = \frac{\Gamma(p+q)}{\Gamma(p)\Gamma(q)} u^{p-1}(1-u)^{q-1} \tag{6}$$

where Γ is the gamma function and the parameters p and q assume the following expressions: $p = \bar{u} / \beta$, $q = (1-\bar{u}) / \beta$ where $\beta = (S_u^2) / (\bar{u}(1-\bar{u}) - S_u^2)$, where $\bar{u}(t) = 1/\Omega \int_{\Omega} u(\mathbf{x},t) dx$ and $S_u^2 = 1/\Omega \int_{\Omega} (u(\mathbf{x},t) - \bar{u}(t))^2 dx$ are the spatial moments of the concentration.

Figure 2 shows the CFDs of a passive solute at the dimensionless time $\tau = 10$ for $\sigma_Y^2 = 0.2, 1, 4$ and 10 .

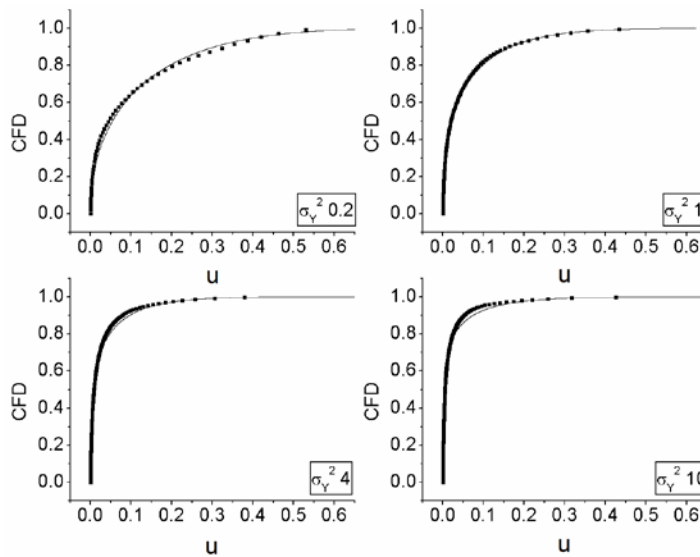


Fig. 2 Spatial CFD of a nonreactive tracer for $\tau = 10$ and $\sigma_Y^2 = 0.2, 1, 4$ and 10 (solid symbols). The Beta model (6) is also shown with a solid line.

Solid squared symbols indicate the CFD of the sample of concentrations obtained numerically with the simulations described above, while the solid line indicates the Beta model obtained by replacing into expressions (6) the spatial concentration moments provided by the numerical simulations.

Inspection of Fig. 2 shows that a larger σ_Y^2 leads to a sharper CFD. The shape of the CFD depends on the stage of the dilution process which has been reached by the plume. At the initial time the dimensionless concentration within the plume is constant and equal to $u = 1$, consequently the CFD is given by $F(\mathcal{U} \leq u) = H(\mathcal{U} - 1)$, where H is the Heaviside step function, i.e. $F(\mathcal{U} \leq u) = 0$ for $u < 1$ and $F(\mathcal{U} \leq u) = 1$ for $u = 1$. Successively, the frequency distributions broaden as an effect of dispersion. However, as time proceeds further the range of concentrations narrows again

as dilution attenuates the concentrations. Figures 1(a) and 2 show that at a given time since injection the plumes are more diluted in the high variance than in the low variance cases. Consequently, in the former case the distribution of the concentration is closer to the concentrations of the ambient water, which is the final condition to which the plumes tend for time tending to infinity.

In all cases we observe a good match between the numerical results and the analytical reference models. This suggests that the Beta distribution model is not limited to weakly heterogeneous formations and can be applied also to high and very high heterogeneous formations provided that a reliable model for the first two spatial concentration moments is available.

CONCLUSIONS

We analysed the evolution of the dilution index and the Cumulative Frequency Distribution of the concentration of an ergodic nonreactive tracer plume transported in heterogeneous formations with σ_Y^2 ranging from 0.2 to 10, i.e. from weakly to highly heterogeneous formations. As expected for a given σ_Y^2 the dilution index increases linearly with time after an initial less than linear behaviour. For a given time the dilution index increases with σ_Y^2 at a rate that decreases with the degree of heterogeneity, showing a weak dependence on σ_Y^2 for $\sigma_Y^2 \geq 4$. A similar behaviour is observed for the maximum concentration. The spatial distribution of the concentration within an ergodic plume is shown to depend on the formation's heterogeneity and is well represented by the Beta model, which depends on the first two spatial concentration moments. These results open interesting perspectives in the application of the stochastic approach in risk assessment, thus contributing to reduce the gap between theory and applications.

Acknowledgement This work has been partially funded by the Italian Ministry of Research and University through the project COFIN 2008 n. 2008A7EBA3.

REFERENCES

- Bellin, A. & Rubin, Y. (1996) Hydro_Gen: A spatially distributed random field generator for correlated properties. *Stoch. Hydrol. Hydraul.* 10(4), 2913–2924.
- Bellin, A. & Tonina, D. (2007) Probability density function of non-reactive solute concentration in heterogeneous porous formations. *Adv. Water Resour.* 94, 109–125.
- Cleary, P. & Monaghan, J. (1999) Conduction modelling using smoothed particle hydrodynamics. *J. Comput. Phys.* 148, 227–264.
- Fiori, A. (2001) The Lagrangian concentration approach for determining dilution in aquifer transport: Theoretical analysis and comparison with field experiments. *Water Resour. Res.* 37(12), 3105–3114.
- Fiori, A. & Dagan, G. (2000) Concentration fluctuations in aquifer transport: a rigorous first-order solution and applications. *J. Contam. Hydrol.* 45(1–2), 139–163.
- Harbaugh, A., Banta, E. R., Hill, M. & McDonald, M. (2000) MODFLOW-2000, the U.S. Geological Survey modular ground-water model - User Guide to modularization concepts and the ground-water flow process. *US Geological Survey Open File Report 00-92*(121p).
- Herrera, P., Massabò, M., & Beckie, R. (2009) A meshless method to simulate solute transport in heterogeneous porous media. *Adv. Water Resour.* 32, 413–429.
- Kapoor, V., Gelhar, L. W. & Miralles-Wilhelm, F. (1997) Bimolecular second-order reactions in spatially varying flows: Segregation induced scale-dependent transformation rates. *Water Resour. Res.* 33(4), 527–536.
- Kitanidis, P. (1994) The concept of dilution index. *Water Resour. Res.* 30(7), 2011–2026.
- LaBolle, E., Fogg, G. & Tompson, A. (1996) Random-walk simulation of transport in heterogeneous porous media: Local mass-conservation problem and implementation methods. *Water Resour. Res.* 32(3), 583–593.
- Monaghan, J. (2005) Smoothed Particle Hydrodynamics. *Rep. Progr. Phys.* 68, 1703–1759.
- Rolle, M., Eberhardt, C., Chiogna, Cirpka, O. A. & Grathwohl, P. (2009), Enhancement of dilution and transverse reactive mixing in porous media: experiments and model based interpretation. *J. Contam. Hydrol.* 110 (3–4), 130–142.
- Rubin, J. (1983) Transport of reacting solutes in porous media: Relation between mathematical nature of problem formulation and chemical nature of reactions. *Water Resour. Res.* 19(5), 1231–1252.
- Tartakovsky, A. M., Meakin, P., Scheibe, T. D. & Eichler West, R. M. (2007) Simulations of reactive transport and precipitation with smoothed particle hydrodynamics. *J. Comput. Phys.* 222, 654–672.
- Tartakovsky, A. M., Redden, G., Lichtner, P. C., Scheibe, T. D. & Meakin, P. (2008) Mixing-induced precipitation: Experimental study and multiscale numerical analysis. *Water Resour. Res.* 44(W06S04).
- Valocchi, A. J., Street, R. L. & Roberts, P. V. (1981) Transport of ion-exchanging solutes in groundwater. Chromatographic theory and field simulation. *Water Resour. Res.* 15(5), 1517–1527.