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On the departure from Gaussianity and Fickianity of transport in highly heterogeneous aquifers

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Abstract The solution of the advective transport problem is difficult because of its intrinsic nonlinearity. The aim of this work is to present a few new results concerning nonlinear transport of passive solutes valid for large conductivity variance. The impact of high heterogeneity is twofold: (i) highly conductive zones may create preferential paths leading to early particle arrival times and large particle displacements; and (ii) the low conductive regions "trap" the solute particles, causing late arrival times and small particle displacements. The combination of the two effects (with the second one prevailing in highly heterogeneous aquifers) leads to continuously increasing values of dispersion coefficients and a departure from the classic Gaussian distribution of trajectories. The effects are larger for the large values of the log-conductivity variance. As a consequence, the transport will seem to be "anomalous" or "non-Fickian" for a long period of time after the injection. Such anomaly is evident, for example, in the tailing of the travel times and the trajectory PDFs.

Keywords Fickianity; Gaussianity; highly heterogeneous formations

INTRODUCTION AND BACKGROUND

Spreading of contaminants in aquifers is caused primarily by the spatial variability of the water velocity $\mathbf{V}(\mathbf{x})$, that in turn stems from heterogeneity in hydraulic conductivity $K(\mathbf{x})$. To account for the seemingly erratic variation by orders of magnitude of K, it is customary to model it as a random space function. Thus, $Y = \ln K$ is usually assumed to be normal and stationary, completely defined by the statistical mean $\langle Y \rangle = \ln K_G$, the variance σ_Y^2 and the two point autocorrelation $\rho(\mathbf{x}_1 - \mathbf{x}_2)$. The latter is characterized by the integral scale $I = \int_0^\infty \rho(x,0,0)\,\mathrm{d}x$. The simplest case of flow is the one which is uniform in the mean, i.e. $\langle \mathbf{V} \rangle = U = const$, pertinent to natural gradient flow in aquifers (mean flow is assumed to be in the x direction).

We consider a finite plume of an inert solute injected in the aquifer at $t = 0$. The contaminant concentration field $C(\mathbf{x},t)$ is also random and it is customary to characterize the plume by its spatial moments: the mass $M = \int C(\mathbf{x},t)\mathrm{d}\mathbf{x}$, the centroid $R = M^{-1}\int xC(\mathbf{x},t)\mathrm{d}\mathbf{x}$, the longitudinal second spatial moment (transverse spreading is not considered here) $S = M^{-1}\int (x-R)^2 C(\mathbf{x},t)\mathrm{d}\mathbf{x}$, the skewness, etc. Deriving the relationship between these spatial moments and the aquifer heterogeneity, by solving the flow and transport equations, has been a topic of intensive research in the last 25 years (see, e.g. monographs by Dagan, 1989, Gelhar, 1993, and Rubin, 2003).

For plumes of large extent with respect to I (ergodic plumes), it was shown that:

$$\frac{\mathrm{d}\langle R\rangle}{\mathrm{d}t} \approx \frac{\mathrm{d}R}{\mathrm{d}t} = \frac{\mathrm{d}\langle X\rangle}{\mathrm{d}t} = U \tag{1}$$

where $X(t;\mathbf{a})$ is the equation of the longitudinal component of the trajectory of a fluid particle that originates at $\mathbf{x} = \mathbf{a}$ at $t = 0$ and $X' = X - \langle X\rangle$ is the fluctuation. Similarly:

$$\frac{\mathrm{d}\langle S\rangle}{\mathrm{d}t} \approx \frac{\mathrm{d}S}{\mathrm{d}t} = \frac{\mathrm{d}X_{11}}{\mathrm{d}t} = 2\alpha_L U \tag{2}$$

where X_{11} is the variance of X, and α_L is the longitudinal macrodispersivity. To investigate longitudinal spreading we refer to the mass of the plume per unit length in the mean flow direction. Furthermore, for the assumed ergodic plume $\overline{C} = \langle\overline{C}\rangle = \int C_0(\mathbf{a}) f(X;\mathbf{a},t)\,\mathrm{d}\mathbf{a}$ where $C_0(\mathbf{a})$ is the concentration of the initial plume and $f(X;\mathbf{a},t)$is the PDF of X at time t of a particle originating at $\mathbf{a}$.

If the displacements X have a Gaussian PDF it can be shown that $\overline{C}$ satisfies the usual advection dispersion equation:

$$\frac{\partial\overline{C}}{\partial t} + U\frac{\partial\overline{C}}{\partial x} = \alpha_L U\frac{\partial^2\overline{C}}{\partial x^2} \tag{3}$$

We shall use the term Gaussian for such transport. Generally, α_L is a function of the travel time of the plume from the injection site (nonlocality). If α_L becomes constant, transport is named Fickian. Otherwise, it is called anomalous. Molecular and pore-scale dispersion are both Gaussian and Fickian.

Many solutions of flow and transport in heterogeneous aquifers were obtained in the past for weak heterogeneity, i.e. for $\sigma_Y^2 < 1$. Solving the equations under a first-order expansion in σ_Y^2 led to the following results (e.g. Dagan, 1984): (i) X is Gaussian at any time if Y is normal, (ii) $f(X;t,\mathbf{a})$ tends asymptotically to Gaussianity for any Y of finite integral scale, and (iii) α_L grows with time during a "setting" period and tends to a constant after a travel distance of say 10 integral scales. This distance is independent of σ_Y^2 and of the statistics of Y.

Elaborate field tests in weakly heterogeneous aquifers (e.g. Mackay *et al.* (1986) for the Borden Site, or LeBlanc *et al.* (1991) for Cape Cod) showed that the field macrodispersivity values are approximately equal to the theoretical ones.

As an alternative, numerical Monte Carlo simulations have been carried out in the past by generating grid values of Y drawn from a multi-Gaussian distribution. The numerical solutions, carried out primarily for 2-D flows (Bellin *et al.*, 1992; Salandin & Fiorotto, 1998) showed that the numerically obtained α_L is close to the first-order approximation for σ_Y^2 as large as unity.

Recently, there has been a flurry of interest in the behaviour of highly heterogeneous formations ($\sigma_Y^2 > 1$). Two topics have attracted attention:

(i) What is the impact of log-conductivity that is not of multi-Gaussian distribution? While elaborate field measurements may provide the univariate PDF of Y and the two point covariance, there are generally not enough data to substantiate the multi-Gaussianity assumption. The latter implies larger integral scales of conductivity classes close to the mean than those pertaining to classes of extreme conductivities. Therefore, one of the ways to check the impact of the multi-Gaussian assumption was to carry out numerical simulations with Y fields in which increased integral scales were attributed to extreme Y values (Wen & Gómez-Hernández, 1998; Zinn & Harvey, 2003).

(ii) How is flow and transport affected by high heterogeneity characterized by large σ_Y^2? This topic is of theoretical interest and relevant to applications (e.g. Boggs *et al.*, 1992) as well. Numerical simulations were carried out for multi-Gaussian structures (Salandin & Fiorotto, 1998; Trefry *et al.*, 2003) besides the aforementioned ones.

These numerical simulations were carried out by conventional methods (FD, FEM) and were limited to 2-D configurations and finite plume sizes, due to the heavy computational burden. Extension to 3-D by these methods seems to be prohibitive at present. We have developed a different numerical approach based on the Analytic Elements Method. (Janković *et al.*, 2003b). In 3-D we modelled the isotropic heterogeneous medium as made up from spherical inclusions embedded in a matrix of conductivity equal to the effective one. The conductivities of the inclusions were drawn from a lognormal distribution. The structure is a heterogeneous medium of a finite integral scale (proportional to the inclusions radii) and a nugget representing the same medium, but with a much smaller integral scale. The great advantage of this model of heterogeneity is that it permits one to achieve highly accurate solutions of flow and transport for high σ_Y^2 (see results for $\sigma_Y^2 = 8$ in the following), for domains and plumes of large size.

It is emphasized that we do not imply that in nature blocks are of spherical size. The model, similar to its other numerical counterparts, mimics (at any desired accuracy) a medium of given log-conductivity PDF and integral scale. Another important point is that the structure is not multi-Gaussian, since the integral scale associated with classes of Y are equal. However, no model of a heterogeneous medium of given univariate PDF and two-point correlation is universal.

The aim here is to present the results of numerical simulations of transport for $\sigma_Y^2 = 4$ and $\sigma_Y^2 = 8$ and to elucidate the two aforementioned aspects: Is transport Gaussian? Is it Fickian?

THE SET-UP AND THE NUMERICAL LABORATORY

The results presented here are based on two numerical simulations. The set up of each simulation, described in Janković *et al.* (2003b), is identical except for the variance of log-conductivity of spherical inclusions ($\sigma_Y^2 = 4$ for the first one and $\sigma_Y^2 = 8$ for the second one) and the background conductivity which was set to the effective conductivity (it equals 1.558 K_G for $\sigma_Y^2 = 4$ and 2.095 K_G for $\sigma_Y^2 = 8$, following Janković *et al.*, 2003a). Inclusions of constant radius R are implanted using periodic packing (face-centred cubic lattice) in a domain shaped as a large prolate ellipsoid with long semi-axis, $0.5L$, equal to twice the short semi-axes. A large number of inclusions

(100 000) with a large volume fraction $n = 0.7$ yielded large L ($L = 166R$) required to achieve ergodicity.

This ensemble of inclusions, subject to uniform flow from infinity in the x direction, behaves like a single inclusion on a large scale. Following the Maxwell solution, the flow inside this large inclusion is uniform and oriented along the x axis regardless of the effective conductivity of the ensemble. Since background conductivity was set to the effective conductivity, the magnitude of the uniform flow inside equals that from infinity.

Particle tracking is conducted in a box $0.55L$ long and $0.2L$ wide and tall (the origin of the coordinate system was set at the centre of the box). The box is smaller than the flow domain; this was required to achieve stationary conditions for particle tracking. 40 000 equally spaced particles are released at the upstream boundary of the box (at $x = -0.275L$) in 200 rows and columns.

RESULTS AND DISCUSSION

This paper aims at analysing longitudinal spread. The database is made up from the values of the particles displacements $X_i(t;\mathbf{a})$ ($i = 1,\ldots,N$) at different t of the $N = 40\ 000$ particles injected at $t = 0$ in the plane $x = -0.275L$. Due to the large, presumably ergodic, plume we can exchange spatial means with ensemble averaging of the displacement of a single particle. A few results relevant to the two topics are discussed here.

Is the plume Gaussian?

In Fig. 1 we have represented the PDF of the longitudinal displacements for the two values $\sigma_Y^2 = 4$ and $\sigma_Y^2 = 8$, centred and normalized with respect to X_{11} at two times. The graphs represent the distribution of the mass per unit length of an ergodic plume at

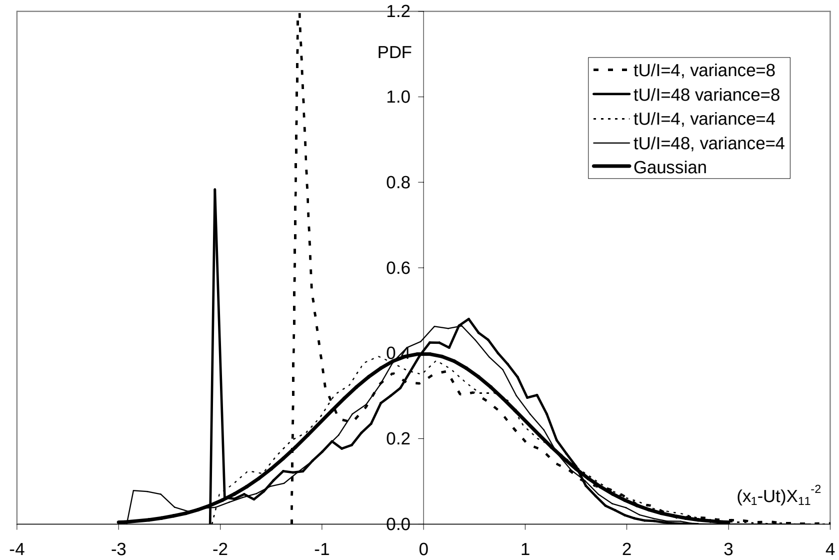

Fig. 1 Distribution $f(X;\mathbf{a},t)$ at two time instances.

different times, normalized by the total injected mass. If the plume were Gaussian, all the graph would have collapsed onto the theoretical Gaussian curve of unit variance.

The striking deviation from the Gaussian shape manifests in the peaks of $f(X;\mathbf{a},t)$ at the injection zone. This effect is related to the low water velocity prevailing in inclusions of low conductivity on one hand, and to the uniformity of instantaneous initial resident concentration along the plane $x = -0.275L$, on the other. It causes solute particles to be trapped for an extended period in this zone. This effect dissipates with time as particles move out of the low K zones. It is also diminished by pore-scale dispersion and molecular diffusion (neglected here) and by injection that is proportional to the flux rather than being uniform. These topics will be discussed in future works. However, we believe that, to a certain extent, this is a phenomenon present in highly heterogeneous formations. The rest of the PDF is slightly skewed with respect to the Gaussian distribution, with an increase of the peak and its advancement ahead of the centre of mass. However, in view of the various assumptions on which the model is based, we do not think that for most practical purposes the departure from Gaussianity of this part of the PDF is important. The picture is similar for the two σ_Y^2 values with augmentation, however, of the mass captured in the regions of low conductivity of the injection zone for $\sigma_Y^2 = 8$.

Is transport Fickian?

We have examined this topic in our previous work (Janković *et al.*, 2003b). Here we present results based on a larger number of particles and extended travel time. The growth of the longitudinal macrodispersivity α_L with time is displayed in a dimensionless form in Fig. 2. α_L was derived by integration of velocity Lagrangean covariance (Janković *et al.*, 2003b) and directly by numerically differentiating the second spatial moment. It is seen that the two agree.

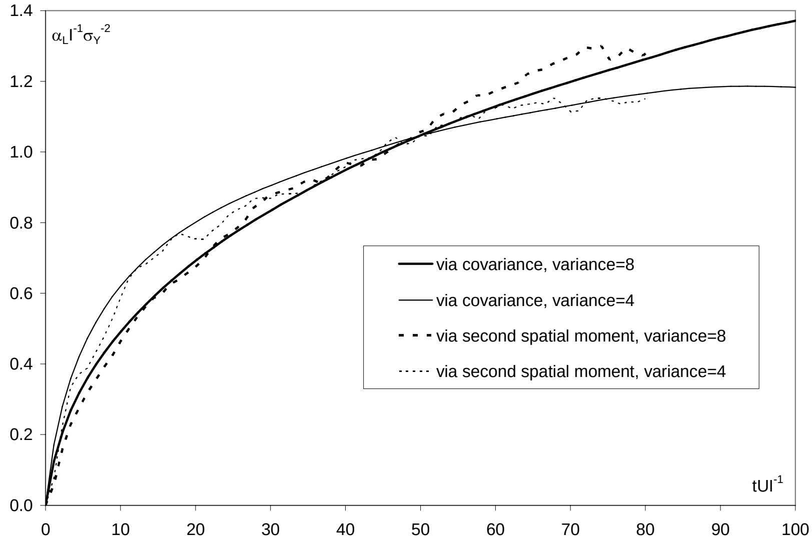

Fig. 2 Dimensionless dispersivity as function of dimensionless time.

If the first-order approximation were valid, the curves for $\sigma_Y^2 = 4$ and $\sigma_Y^2 = 8$ should have collapsed on a unique one. The difference in the magnitude of α_L at a given travel time reflects the impact of nonlinearity, which is seen to be moderate in the time interval considered here. The more serious departure from the linear solution concerns the "setting" time. It is seen that transport becomes Fickian only for $tUI^{-1} > 100$ for $\sigma_Y^2 = 4$ and α_L does not reach a plateau for $\sigma_Y^2 = 8$ at this large time. We know that eventually transport has to become Fickian for any σ_Y^2, but the tendency is very slow. Therefore, for an observer that monitors the plume at shorter travel distances, transport appears to be anomalous.

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