

Solute transport in evolving scale formations: analysis of the concentration field

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Abstract We compare analytical solutions for the expected value of the absolute and relative concentrations of a conservative solute in evolving scale formations. The spatial variability of the logarithm of hydraulic conductivity is described by using a self-similar random space function with a suitable cut-off length representing the physical constraints imposed by geological boundaries. A large disparity is observed between the concentrations predicted by using the two approaches with the absolute concentration showing the faster decreasing rate with time. In the relative approach, large-scale variations of the hydraulic conductivity do not contribute to the spreading of the solute and the plume is more compact. We conclude that the absolute approach underestimates concentration levels at the accessible environment.

INTRODUCTION

Recent theoretical studies suggest that natural formations exhibit a hierarchy of characteristic scales of heterogeneity. Although this assumption is currently under debate and not yet confirmed by conclusive experimental findings, it has been used by several authors to develop models of the hydraulic property variations in the subsurface (e.g. Wheatcraft & Tyler, 1988; Neuman, 1990; Glimm & Sharp, 1991; Hassan *et al.*, 1997).

A commonly used multiscale geostatistical model of spatial variability assumes that the logarithm of hydraulic conductivity $Y = \ln K$ is a self-similar random space function characterized by the following semivariogram:

$$\gamma_Y(r) = \frac{1}{2} \langle [Y(x+r) - Y(x)]^2 \rangle = a|r|^\beta \quad (1)$$

where r is the separation distance, $\beta = 2H$, where H is the Hurst coefficient (Feder, 1988), and a is a suitable constant. The geological formation described by the model in equation (1) is without a characteristic scale of variability and both the variance and the integral scale of the log of hydraulic conductivity grow unbounded with the observation scale. One of the criticisms to this model is that it assumes the existence of an infinite hierarchy of scales of variability, while in natural formations geological boundaries pose an upper limit, which is structural, to the underlying geological processes that contributed to the formation of the aquifer (Anderson, 1997). This

problem is bypassed by the introduction of the “cut-off” length L_{\max} , which is defined as the maximum scale of variability existing in the aquifer (e.g. Kumbhkar & Wen, 1993; Dagan, 1994; Zhan & Wheatcraft, 1996). Once the cut-off is imposed, Y becomes a stationary random space function characterized by the constant mean $\langle Y \rangle$ and variance $\sigma_Y^2(L_{\max}) = aL_{\max}^\beta$, and the following covariance function:

$$\begin{cases} C_Y(r) = \sigma_Y^2 - a|r|^\beta & (r \leq L_{\max}) \\ C_Y(r) = 0 & (r > L_{\max}) \end{cases} \quad (2)$$

Variations of the log of hydraulic conductivity of the type described by equation (2) lead to a dispersion coefficient that grows with time reaching the constant Fickian limit for $tU/L_{\max} \gg 1$. In practice this condition is hardly obtained since L_{\max} is typically larger than the domain of interest. Thus, in this work we analyse the case $tU/L_{\max} < 1$ which is relevant for practical applications.

THEORETICAL BACKGROUND

We consider the transport of a conservative solute in a two-dimensional formation with the variations of the log of hydraulic conductivity described by equation (2). At the time $t = 0$, a conservative solute with concentration $C_0(\mathbf{a})$ is released over the area A_0 . Neglecting pore-scale dispersion and assuming the porosity as constant, the concentration of the solute assumes the following expression (Dagan, 1982):

$$C(\mathbf{x}, t) = \int_{A_0} C_0(\mathbf{a}) \delta(\mathbf{x} - \mathbf{X}) d\mathbf{a} \quad (3)$$

where $\mathbf{X} = \mathbf{X}(t; \mathbf{a})$ is the trajectory of the particle injected at $\mathbf{x} = \mathbf{a}$. The expected value of the concentration is given by:

$$\langle C(\mathbf{x}, t) \rangle = \int_{A_0} C_0(\mathbf{a}) f_X(\mathbf{x}; t, \mathbf{a}) d\mathbf{a} \quad (4)$$

where $f_X(\mathbf{X})$ is the trajectory's probability density function (pdf) of the particle injected at the location $\mathbf{x} = \mathbf{a}$.

Recent studies showed that the large scale hydraulic property variations cause the plume to wander as a whole while the spreading of the solute is controlled by small scale heterogeneities with the upper limit imposed by the plume size (e.g. Kitanidis, 1988; Dagan, 1991). Thus, the absolute concentration in equation (4) applies when the source size is much larger than L_{\max} . In the attempt to remove the effect of plume meandering, Fiori (2000) introduced the concept of relative concentration:

$$C_r(\xi, t) = \int_{A_0} C_0(\mathbf{a}) \delta(\xi - \mathbf{W}) d\mathbf{a} \quad (5)$$

where $\xi = \mathbf{x} - \mathbf{R}(t)$ is the coordinate system centred on the trajectory of the plume's centroid $\mathbf{R}(t)$ and $\mathbf{W} = \mathbf{X}(t) - \mathbf{R}(t)$. Andricevic & Cvetkovic (1999) proposed a similar approach for the solute flux. Thus, the expected value of C_r assumes the following expression:

$$\langle C_r(\xi, t) \rangle = \int_{A_0} C_0(\mathbf{a}) f_W(\xi; t, \mathbf{a}) d\mathbf{a} \quad (6)$$

where $f_W(\mathbf{W})$ is the pdf of the trajectory of the particle injected at \mathbf{a} , relative to the trajectory of the plume's centroid.

EXAMPLE OF APPLICATION AND DISCUSSION OF THE RESULTS

We consider an instantaneous release of a conservative solute with constant concentration C_0 over the rectangular area A_0 where longitudinal and transverse sides are l_1 and l , respectively. The mean velocity U is aligned with the x_1 direction and the source is centred at the origin of the coordinate system (x_1, x_2) . In the following, the subscripts 1 and 2 refer to the longitudinal and transverse directions, respectively.

At the first order of approximation in σ_Y^2 , both f_X and f_W are multivariate normal (Dagan, 1982) and characterized by the following statistical moments:

$$\langle \mathbf{X} \rangle = \mathbf{a} + \mathbf{U}t \quad X_{ij}(t) = \langle X'_i(t; \mathbf{a}) X'_j(t; \mathbf{a}) \rangle \quad (7)$$

$$\langle \mathbf{W} \rangle = \mathbf{a} \quad W_{ij}(t; \mathbf{a}) = \langle W'_i(t; \mathbf{a}) W'_j(t; \mathbf{a}) \rangle = X_{ij}(t) + R_{ij}(t) - 2O_{ij}(t; \mathbf{a}) \quad (8)$$

where X'_i and W'_j are the fluctuations of the absolute and relative particle trajectories from the corresponding mean values $\langle X_i \rangle$ and $\langle W_i \rangle$. In addition, $R_{ij} = \langle R'_i R'_j \rangle$ ($i, j = 1, 2$) are the second-order moments of the residual displacement of the plume's centroid, and $O_{ij} = \langle X'_i R'_j \rangle$ ($i, j = 1, 2$) represent the cross-correlations between the residual displacements of the particle and the plume's centroid trajectory. The structure of equation (8) suggests that at the first-order of approximation, W_{ij} is independent of the imposed cut-off, which instead affects X_{ij} . This result is consistent with the non-ergodic spatial moments analysis performed by Dagan (1994) and Bellin *et al.* (1996).

After these preparatory steps the mean absolute concentration $\langle C \rangle$ assumes the following expression:

$$\frac{\langle C(\mathbf{x}, t) \rangle}{C_0} = \frac{1}{4} \left\{ \operatorname{erf} \left[\frac{x_1 - L + l_1/2}{\sqrt{2X_{11}(t)}} \right] - \operatorname{erf} \left[\frac{x_1 - L - l_1/2}{\sqrt{2X_{11}(t)}} \right] \right\} \cdot \left\{ \operatorname{erf} \left[\frac{x_2 + l/2}{\sqrt{2X_{22}(t)}} \right] - \operatorname{erf} \left[\frac{x_2 - l/2}{\sqrt{2X_{22}(t)}} \right] \right\} \quad (9)$$

where $L = Ut$ is the travel distance; the moments X_{11} , X_{22} are independent of \mathbf{a} .

Closed form solutions of $\langle C_r \rangle$ can not be obtained in the general case. Seeking for a simplified solution, we observe that for $l_1 \ll l$ the trajectories of two particles released at the same transverse coordinate can be assumed fully correlated such that W_{ij} vanishes for $i \neq j$, and the moments O_{ii} , $i = 1, 2$ are independent from a_1 . Thus, the multivariate normal pdf f_W reduces to the product of two independent univariate

normal pdfs:

$$f_W(\xi) = \frac{1}{2\pi\sqrt{W_{11}(t;a_2)W_{22}(t;a_2)}} \exp\left[-\frac{(\xi_1 - a_1)^2}{2W_{11}(t;a_2)}\right] \exp\left[-\frac{(\xi_2 - a_2)^2}{2W_{22}(t;a_2)}\right] \quad (10)$$

With the above assumption, $\langle C_r \rangle$ equation (6) assumes the following expression:

$$\frac{\langle C_r(\xi, t) \rangle}{C_0} = \frac{1}{2\sqrt{2\pi}} \int_{-1/2}^{1/2} \exp\left[-\frac{(\xi_2 - a_2)^2}{2W_{22}(t;a_2)}\right] \left\{ \operatorname{erf}\left[\frac{\xi_1 + l_1/2}{\sqrt{2W_{11}(t;a_2)}}\right] - \operatorname{erf}\left[\frac{\xi_1 - l_1/2}{\sqrt{2W_{11}(t;a_2)}}\right] \right\} da_2 \quad (11)$$

where a numerical quadrature is needed. Thus, the expected values of the absolute and relative concentrations can be computed by substituting in equations (9) and (11) the first-order expressions of X_{ii} and W_{ii} , respectively. Figure 1(a) shows $\langle C_r \rangle / C_0$ and $\langle C \rangle / C_0$ at $\mathbf{x} = \langle \mathbf{R}(t) \rangle$ versus the mean travel distance $L = Ut$, and for $\beta = 0.5$ ($H = 0.25$). The expected value of the absolute concentration is shown for two different cut-off lengths. Furthermore, $al^\beta = 0.01$ such that the variance of the log conductivity field is fixed at 0.032 and 0.045 for $L_{\max}/l = 10$ and 20, respectively.

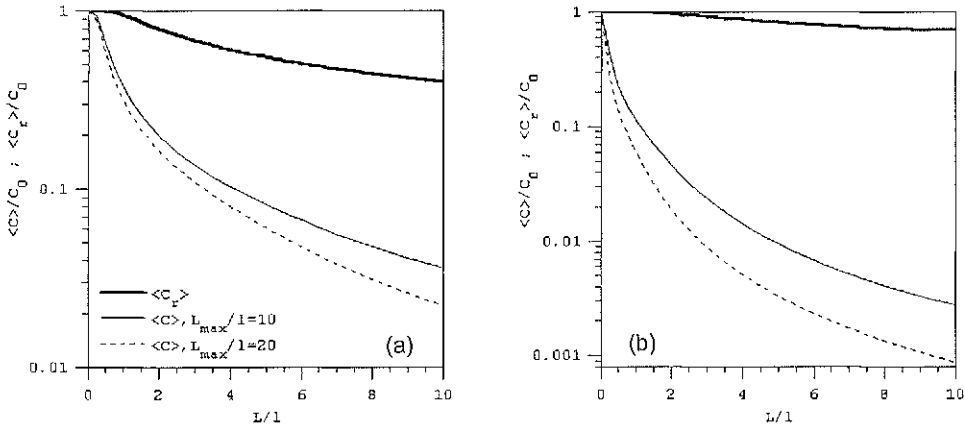


Fig. 1 Absolute $\langle C \rangle$ equation (9) and relative $\langle C_r \rangle$ equation (11) mean concentrations versus $L/l = Ut/l$, for two values of L_{\max} : (a) $\beta = 0.5$, and (b) $\beta = 1.5$. In all cases $l_1/l = 0.1$ and $al^\beta = 0.01$.

Inspection of Fig. 1(a) reveals that a larger L_{\max} results in a faster decay of $\langle C \rangle$ with the travel distance. This is consistent with the fact that the variance of the hydraulic conductivity increases with the imposed cut-off length leading to a larger spreading of the solute. The expected value of C_r , which is independent from L_{\max} , is much larger than $\langle C \rangle$ with differences that exceed one order of magnitude for $L/l > 5$. The plume is more compact than in the absolute concentration approach since the spreading of the solute is affected only by variations of Y acting at scales smaller than the source size, while variations occurring at larger scales cause the plume to wander as a whole.

Therefore, in the relative concentration approach the large-scale variations of Y do not introduce additional solute spreading such that $\langle C \rangle$ is expected to resemble the actual concentration much better than $\langle C \rangle$, which in turn is affected by variations of Y acting at scales smaller than L_{\max} . Figure 1(b) shows the case $\beta = 1.5$ ($H = 0.75$). The log-conductivity variance is fixed at 0.32 and 0.89 for $L_{\max}/l = 10$ and 20, respectively. The larger log-conductivity variance results in a much faster decreasing rate of $\langle C \rangle$. Conversely, the relative concentration shows a smaller attenuation with the travel distance. This is due to the disparity existing between the scales of the dominant variations of the hydraulic conductivity in the cases shown in Figs 1(a) and 1(b). For $\beta < 1$ the convexity of the semivariogram is downward such that the variations of the log of hydraulic conductivity are stronger at small scales while the opposite is observed for $\beta > 1$. In addition, the results shown in Figs 1(a) and 1(b) are for the same value of $\sigma_Y^2(l)$. Considering that solute spreading is caused by permeability variations of scales smaller than the source size, one can conclude that for a given $\sigma_Y^2(l)$, the larger spreading is observed for $\beta < 0.5$. On the other hand, for $\beta > 1$ the plume experiences a stronger meandering while it remains more compact than for $\beta < 1$, since the small scale variations of the hydraulic conductivity are weaker. For $L > L_{\max}$ the cut-off length L_{\max} affects the spreading of the solute, since the plume samples variations of the hydraulic property at all the relevant scales.

Figures 2(a) and 2(b) shows R_{11} and R_{22} , versus the travel distance L . For a given travel time R_{11} is one order of magnitude larger than R_{22} and both increase with the travel distance. Furthermore, a larger cut-off length results in larger values of both R_{11} and R_{22} , since variations of the log of hydraulic conductivity are much stronger at all the scales of heterogeneity. As a further confirmation of what was observed in Figs 1(a) and 1(b), a larger β results in a stronger plume meandering and a proportionally smaller solute spread as can be argued from the larger values of R_{11} and R_{22} observed in Figs 2(a) and 2(b). The large overestimation of the attenuation of the solute

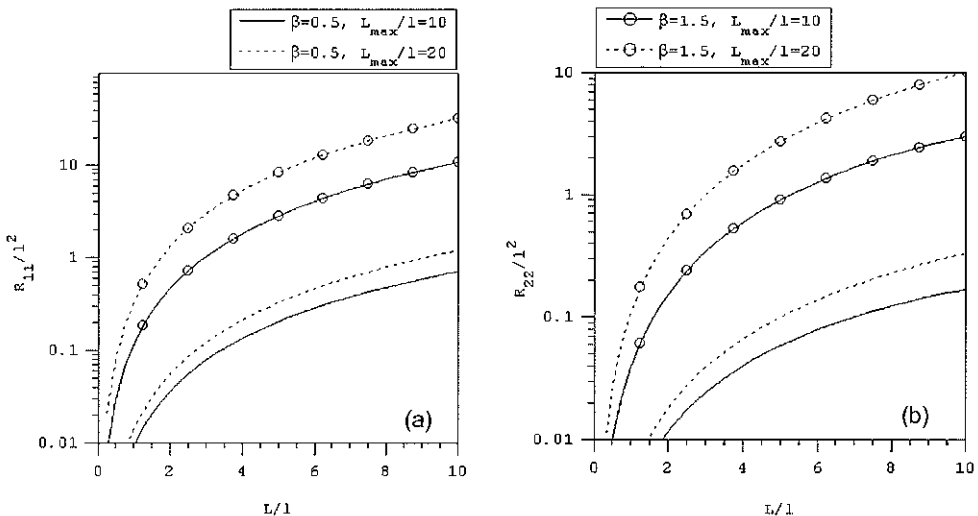


Fig. 2 The longitudinal R_{11} (a), and transverse R_{22} (b) second moments of the plume centre of mass as a function of $L/l = Ut/l$, for a few values of the cut-off L_{\max} ; $a^{\beta} = 0.01$; solid lines: $L_{\max}/l = 10$, dashed lines: $L_{\max}/l = 20$; circles: $\beta = 1.5$.

concentration obtained employing the absolute concentration concept is of some concern for practical applications where the environmental regulations limit the maximum concentration of the contaminants in the accessible environment.

Furthermore, we note that the relative concentration concept is representative of the effective plume dispersion, since it selects actively the variations of the hydraulic property that contribute effectively to the spreading of the solute.

We can therefore conclude that the absolute concentration approach shows a tendency to overestimate the spreading of a conservative solute such that the predicted concentrations may be an order of magnitude smaller than the actual concentration. On the other hand, the relative concentration approach filters the large-scale hydraulic conductivity variations leading to a smaller solute spread and a larger concentration. However the relative concentration approach is affected by the uncertainty in the spatial variability of the hydraulic conductivity at scales smaller than l , which can be reduced by conditioning with measurements.

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