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A theoretical framework for modeling dilution enhancement of non-reactive solutes in heterogeneous porous media

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ABSTRACT

Spatial heterogeneity of the hydraulic properties of geological porous formations leads to erratically shaped solute clouds, thus increasing the edge area of the solute body and augmenting the dilution rate. In this study, we provide a theoretical framework to quantify dilution of a non-reactive solute within a steady state flow as affected by the spatial variability of the hydraulic conductivity. Embracing the Lagrangian concentration framework, we obtain explicit semi-analytical expressions for the dilution index as a function of the structural parameters of the random hydraulic conductivity field, under the assumptions of uniform-in-the-average flow, small injection source and weak-to-mild heterogeneity. Results show how the dilution enhancement of the solute cloud is strongly dependent on both the statistical anisotropy ratio and the heterogeneity level of the porous medium. The explicit semi-analytical solution also captures the temporal evolution of the dilution rate; for the early- and late-time limits, the proposed solution recovers previous results from the literature, while at intermediate times it reflects the increasing interplay between large-scale advection and local-scale dispersion. The performance of the theoretical framework is verified with high resolution numerical results and successfully tested against the Cape Cod field data.

1. Introduction

Quantifying dilution of contaminant clouds in flows within natural geological (porous) formations is important in many hydrological, environmental and reservoir engineering applications. Dilution plays a key role in defining (bio)remediation strategies (Cirpka and Valocchi, 2007; Kitanidis, 2012), attenuation of pollutant concentration levels at field and catchment scales (Fiori and Dagan, 2000; Rinaldo et al., 2011) and reduction of human health risk (de Barros et al., 2009). The dilution rate of a contaminant traveling in a porous medium depends on the complex interaction between the heterogeneous distribution of the hydraulic conductivity and the intensity of diffusion and local-scale dispersion mechanisms. Variability in the hydraulic conductivity (and consequently, in the velocity field), leads to an irregular spreading of the cloud, which increases the surface area between the solute and the surrounding fluid. As a consequence of heterogeneity, the dilution of the cloud is enhanced (Dentz et al., 2011; Kapoor and Kitanidis, 1998). Therefore, to improve the predictive capabilities of solute transport models in heterogeneous porous formations, it is essential to reproduce, as close as possible, the complex spatial patterns of the hydraulic conductivity at the field-scale onto transport while accounting for the uncertainty stemming from the lack of a detailed site characterization. For such reasons stochastic methods are appealing when dealing with transport of solutes clouds at the field-scale, because they allow to treat uncertainty and spatial variability within a unified framework (Rubin, 2003).

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Along this line, Fiori and Dagan (2000) and Tonina and Bellin (2008) illustrated how the statistical moments of local concentration are affected by the structural parameters of the random conductivity field and the size of the sampling device. These authors, amongst others (Kapoor and Gelhar, 1994; Kapoor and Kitanidis, 1998; Pannone and Kitanidis, 1999; Vanderborght, 2001; Zhang and Neuman, 1996), showed how the concentration variance is destroyed by diffusion and local-scale dispersion. The joint impact of heterogeneity and local-scale dispersion on the concentration probability density function is also reported (Cirpka et al., 2008; Dentz and Tartakovsky, 2010; de Barros and Fiori, 2014). The significance of heterogeneity on dilution enhancement due to transverse mixing can be found in experiments at the laboratory scale (Bauer et al., 2008; Rolle et al., 2009) and through modeling (Cirpka et al., 2011).

With the aim of globally quantifying dilution of solute clouds in the subsurface environment, Kitanidis (1994) proposed an entropy-based metric denoted as the dilution index. Thierrin and Kitanidis (1994) demonstrated the utility of the dilution index as a global indicator of dilution by evaluating its temporal evolution for the idealized case of transport in a homogeneous velocity field and for the intensively monitored clouds in field-scale transport experiments. A critical dilution index was quantified to address mixing of reactive two-dimensional steady-state clouds (Chiogna et al., 2011). Within the context of flow topology, de Barros et al. (2012) used the dilution index to illustrate how local flow topological features control the dilution growth rate of a solute blob. Impact of heterogeneity on other measures of dilution, such as the scalar dissipation rate, can be found in the literature (e.g., de Dreuzy et al., 2012; Le Borgne et al., 2010 and references therein). Despite the great deal of attention given to this subject, there is still a compelling need to improve our fundamental understanding of how the spatial variability of the conductivity impacts dilution and mixing in groundwater.

This work aims at developing a theoretical framework that predicts dilution of solutes (in both two- and three-dimensional heterogeneous flows) while establishing a link between the concentration field and the hydrogeological properties of the formation, thereby providing physical insight of the key parameters controlling the dilution rate of the solute cloud. In particular, we establish a closed-form functional relationship between the dilution index and the solute particle trajectories, which depends on the structural parameters that characterize the spatial variability of the hydraulic conductivity. The proposed methodology is based on the Lagrangian concentration concept introduced by Fiori (2001). The appealing feature of this framework is that it automatically filters out uncertainty in local concentration values.

Our results highlight the importance of both the statistical anisotropy ratio of the medium (e.g., ratio between the conductivity correlation scales) and the log-conductivity variance in controlling the dilution state of the solute and its temporal scaling. The semi-analytical results are compared to two-dimensional, high resolution, numerical simulations. Finally, we successfully test the performance of the proposed modeling framework against field data from the Cape Cod (Massachusetts, USA) experiment (Leblanc et al., 1991) by revisiting the experimental analysis of Thierrin and Kitanidis (1994). Our semi-analytical solution is developed for small injection sources with respect to the heterogeneity correlation scale (e.g., point-like injection sources) using perturbation theory therefore valid for weak-to-mild heterogeneity.

2. Problem formulation

We consider an incompressible steady-state flow field through a fully saturated heterogeneous porous medium with a spatially heterogeneous locally isotropic hydraulic conductivity $K(\mathrm{x})$ and constant porosity ϕ. As a consequence, the resulting Darcy-scale velocity field $\mathbf{u}(\mathbf{x})$ is spatially variable and referenced through a Cartesian coordinate system: $\mathbf{x}(x_1,\ldots x_N)$ where N is the space dimensionality. The flow field under consideration is far from boundary effects and uniform-in-the-average with mean velocity $\langle \mathbf{u} \rangle \equiv \mathbf{U}(U_1, 0, 0)$. The governing flow equation is given by

$$\nabla \cdot [K(\mathbf{x})\nabla h(\mathbf{x})] = 0 \quad (1)$$

where h is the hydraulic head and the velocity field is determined by Darcy's law $\mathbf{u}(\mathbf{x}) = -[K(\mathbf{x}) / \phi]\nabla h$.

The spatial structure of the flow field is governed by log-conductivity $Y = \ln K$ field which is assumed to be a statistically stationary *Random Space Function* (RSF), normally distributed with mean $\langle Y \rangle$ and variance σ_Y^2. In this work, $\langle \cdot \rangle$ denotes ensemble expectation. The statistically stationary covariance function of Y is given by $C_Y(|\mathbf{x} - \mathbf{x}'|)$ and is fully characterized by the integral scales $I_{Y,i}$ (with $i = 1,\ldots,N$) in the N principal directions, and the log-conductivity variance σ_Y^2. Hereafter, for simplicity of notation, all the RSF parameters are summarized within the vector $\boldsymbol{\Theta} = [\langle Y \rangle, \sigma_Y^2, I_{Y,1}, \ldots, I_{Y,N}]$.

A solute with initial uniform Concentration C_o is instantaneously injected into the aquifer. The injection zone is characterized by a parallelepiped of volume $\mathcal{V}_o = \prod_{i=1}^{N} L_i$. Transport is governed by the advection–dispersion equation

$$\frac{\partial C(\boldsymbol{x}, t)}{\partial t} = \mathbf{D}_d \nabla^2 C(\boldsymbol{x}, t) - \mathbf{u}(\boldsymbol{x}) \cdot \nabla C(\boldsymbol{x}, t), \quad (2)$$

with the following initial condition

$$C(\mathbf{x}, 0) = \begin{cases} C_o & \forall \mathbf{x} \in \mathcal{V}_o \\ 0 & \forall \mathbf{x} \notin \mathcal{V}_o \end{cases} \quad (3)$$

where C is the local (resident) concentration, t denotes time and $\mathbf{D}_\mathrm{d}$ is the local-scale dispersion tensor. For the purpose of this work, we assume $\mathbf{D}_\mathrm{d}$ to be constant. Because $Y = \ln K$ is a RSF, the local concentration C is also regarded as a random function.

3. The Lagrangian approach to transport in heterogeneous formations

In the following, we will make use of the Lagrangian framework to quantify $C(\boldsymbol{x}, t)$ within the spatially variable conductivity field which obeys Eq. (2) (Fiori and Dagan, 2000)

$$\begin{aligned} C(\boldsymbol{x}, t) &= C_0 \int_{\mathcal{V}_0} \delta[\mathrm{x} - \mathbf{X}_t(t; \mathbf{a})] d\mathbf{a} \\ &= C_0 \int_{\mathcal{V}_0} \delta\left[\mathrm{x} - \mathbf{a} - \mathbf{U}t - \mathbf{X}'(t; \mathbf{a}) - \mathbf{X}_d\right] d\mathbf{a} \end{aligned} \quad (4)$$

where δ is the Dirac delta function and $\mathbf{a}$ denotes the initial location of a solute particle within source injection volume