



Stochastic analytical modeling of the biodegradation of steady plumes

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ABSTRACT

We present a stochastic analytical framework to assess the contaminant concentration of a steady plume undergoing biodegradation. The method is focused on heterogeneous formations, and it embeds both fringe and core degradation. The Lagrangian concentration approach of Fiori (2001) was employed, which is suited for describing the interplay between the large scale advection caused by heterogeneity and the local dispersion processes. The principal scope of the model is to provide a relatively simple tool for a quick assessment of the contamination level in aquifers, as function of a few relevant, physically based dimensionless parameters. The solution of the analytical model is relatively simple and generalizes previous approaches developed for homogeneous formations. It is found that heterogeneity generally enhances mixing and degradation; in fact, the plume shear and distortion operated by the complex, heterogeneous velocity field facilitates local dispersion in diluting the contaminant and mixing it with the electron acceptor. The decay of the electron donor concentration, and so the plume length, is proportional to the transverse pore-scale dispersivity, which is indeed the parameter ruling mixing and hence degradation. While the theoretical plume length is controlled by the fringe processes, the core degradation may determine a significant decay of concentration along the mean flow direction, thus affecting the length of the plume. The method is applied to the crude oil contamination event at the Bemijdi site, Minnesota (USA).

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1. Introduction

Many contaminants present in the subsurface can be degraded by biological processes where microorganisms, naturally present in the groundwater, metabolize organic contaminant producing inorganic material (Farhadian et al., 2008). Natural degradation is thus an attractive technique for its effects and costs, although it may involve long times and large areas. In the degradation processes the contaminant acts as the electron donor and carbon source, and degradation occurs when the contaminant mixes with an electron acceptor, such as oxygen or nitrate. Hence, local mixing is a crucial component which triggers the biodegradation of organic compounds in groundwater. In turn, mixing is enhanced by the spatial heterogeneity of the aquifer hydraulic properties. In

fact, solute plumes do stretch and distort because of heterogeneity, developing solute “fingers” with significant lateral concentration gradients, such that transverse local dispersion/diffusion is more effective in diluting the contaminant and mixing the electron donor and acceptor. Hence, the interplay between local dispersion and large scale advection due to heterogeneity is crucial in determining local mixing and thus the degradation processes.

Plumes originating from old contamination events may easily reach steady state conditions; previous analyses dealing with field investigation (e.g. Essaid et al., 1995; Maier and Grathwohl, 2005; Van Breukelen et al., 2004) suggest adopting the steady state transport to model the key features of the transport processes in such environments. Most of the previous studies on the subject have analyzed steady transport in homogeneous formations (e.g. Cirpka and Valocchi, 2007; Domenico, 1987; Gutierrez-Neri et al., 2009; Liedl et al., 2005), and only a limited number of papers have considered reactive

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transport in heterogeneous media, and mainly for transient plumes (e.g. Bellin et al., 2011; Dagan and Cvetkovic, 1996; Severino et al., 2012).

In the present work we develop analytical solutions for steady transport of a contaminant undergoing biodegradation. We are interested in relatively simple solutions, with a limited number of parameters, to be used as a screening tool for a rapid analysis of the area subject to contamination and for risk assessment studies. Following a customary approach (e.g. Dagan, 1989; Rubin, 2003) we model the hydraulic conductivity K as space random function with given statistical properties. Under steady conditions, transport is ruled by the lateral and vertical spreading, and longitudinal transport can be assumed as deterministic (Cirpka and Valocchi, 2007; Zarlunga and Fiori, 2013a).

The microbial reactions are separated in two components, pertaining to fringe and core biodegradation, respectively (e.g. Gutierrez-Neri et al., 2009). The former refers to the aerobic processes (e.g. denitrification and sulfate reduction) occurring at the plume fringe (Davis et al., 1999), while the latter concerns with anaerobic processes (e.g. iron and manganese reduction and methanogenesis; see Essaid et al., 1995) and mainly occurs at the core of the plume. For the fringe processes, the microbial reaction is assumed as instantaneous, similar to the approach of Borden and Bedient (1986) and Ham et al. (2004); the assumption of instantaneous reaction is supported by the numerical investigations presented by Chu et al. (2005), Cirpka and Valocchi (2007), Maier and Grathwohl (2005), which show that the limiting factor for natural attenuation is the vertical and transversal mixing rather than the reaction kinetics. However, the adopted approximations can be justified considering the uncertainty on the site characterization and on the biological processes involved. In turn, a simple first-order kinetics is adopted for the core degradation processes (Domenico, 1987; Gutierrez-Neri et al., 2009).

Flow and transport are solved by the First order approximation approach (Dagan, 1989; Fiori, 1996; Rubin, 2003), which is formally valid for low/moderate heterogeneity. Along the stochastic approach, all the flow and transport variables are considered as space random functions. The Lagrangian concentration definition of Fiori (2001) is employed here; its main scope is to filter out the advective components which do not directly contribute to mixing, and thus focus on the local mixing processes which are the main triggers of degradation. The assumptions adopted allow developing relatively simple analytical or semianalytical solutions for steady transport undergoing biodegradation in heterogeneous porous formations.

2. Mathematical framework

Steady flow of mean uniform velocity U takes place in a natural, heterogeneous porous formation, with spatially variable hydraulic conductivity $K(\mathbf{x})$, where $\mathbf{x}(x_1, x_2, x_3)$ is the reference system aligned with the mean flow direction. The logconductivity $Y = \ln K$ is modeled as a stationary spatial random variable, normally distributed, with mean and variance equal to $\langle Y \rangle$ and σ_Y^2 , respectively. The spatial correlation is described by the two-point covariance $C_Y(\mathbf{r})$, which depends on the vector distance \mathbf{r} between two points. As suggested by field evidence (see e.g. Table 2.3 in Rubin, 2003), C_Y is assumed as axisymmetric, with integral scales equal to $(l_{Y,h}, l_{Y,h}, l_{Y,v})$ along

the directions (x_1, x_2, x_3) ; thus, the mean velocity \mathbf{U} is aligned with the principal anisotropy direction $l_{Y,h}$. With $e = l_{Y,v}/l_{Y,h}$ the anisotropy ratio, it is usually $e = O(10^{-1})$ (Rubin, 2003). The velocity field \mathbf{v} in the medium is random and spatially variable, with mean equal to $\langle \mathbf{v} \rangle = (U, 0, 0)$.

A contaminant (e.g. an organic compound which serves as an electron donor) is continuously released over the area $A_0 = L_2 L_3$ placed at $x_1 = 0$, with $L_2 L_3$ the transverse and vertical dimensions of the plume, respectively. The contaminant concentration C_D over the injection area is constant and equal to C_D^0 . Solute undergoes large scale macrodispersion and pore-scale dispersion (hereinafter PSD), and in the large time limit the concentration assumes a steady profile. While for transient plumes the longitudinal dispersion–diffusion plays a crucial role, steady transport is mainly governed by the lateral and vertical macro- and pore-scale dispersion (e.g. Wexler, 1992; Zarlunga and Fiori, 2013b).

After the contaminant release, complex biological processes occur; if all the substrates involved are available, natural communities of microorganisms (bacteria and fungi) mineralize organic C, N, P, S and CO_2 or other inorganic compounds which are released in the groundwater. The previous reactions constitute the natural attenuation process or bioremediation. Microorganism present in the subsurface are highly versatile, and biodegradation occurs in many different field conditions. We schematize the complex reactions in two categories, whether degradation occurs under aerobic or anaerobic conditions, respectively. This assumption is rather common, (e.g. Essaid et al., 2011; Gutierrez-Neri et al., 2009; Suthersan and Payne, 2005) and allows to grasp the main features occurring in the natural attenuation processes.

Fringe reactions account for the contaminant degradation in aerobic conditions. Those reactions typically take place on the plume fringe where the contaminant, acting as electron donor (subscript D), is mixed with the electron acceptor (subscript A) provided by the fresh, uncontaminated water. Because of the relatively fast kinetics (Chu et al., 2005; Davis et al., 1999) the pore scale dispersion plays a crucial role in controlling the reactions. The latter are summarized through a global bimolecular irreversible reaction



where P is the product and f_i is the stoichiometric coefficient of the i -th specie.

Then, *core reactions* account for the reactions occurring in anoxic conditions, typically in the core plume region (e.g. Fe, Mn reduction and methanogenesis). Although the core reactions are often neglected in many degradation models, recent studies recognize the primary importance of those mechanism on the plume degradation and the need to introduce them, in combination with the fringe model (Amos et al., 2011; Essaid et al., 2011; Lovley et al., 1989). Those reactions are usually represented by the exponential model

$$\frac{dC_D}{dt} = -\lambda C_D \quad (2)$$

Each contaminant species is characterized by its own decay coefficient that depends on the particular field condition; for simplicity a global single effective coefficient λ is adopted here.

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