



Exposure-time based modeling of nonlinear reactive transport in porous media subject to physical and geochemical heterogeneity



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ABSTRACT

Transport of reactive solutes in groundwater is affected by physical and chemical heterogeneity of the porous medium, leading to complex spatio-temporal patterns of concentrations and reaction rates. For certain cases of bioreactive transport, it could be shown that the concentrations of reactive constituents in multi-dimensional domains are approximately aligned with isochrones, that is, lines of identical travel time, provided that the chemical properties of the matrix are uniform. We extend this concept to combined physical and chemical heterogeneity by additionally considering the time that a water parcel has been exposed to reactive materials, the so-called exposure time. We simulate bioreactive transport in a one-dimensional domain as function of time and exposure time, rather than space. Subsequently, we map the concentrations to multi-dimensional heterogeneous domains by means of the mean exposure time at each location in the multi-dimensional domain. Differences in travel and exposure time at a given location are accounted for as time difference. This approximation simplifies reactive-transport simulations significantly under conditions of steady-state flow when reactions are restricted to specific locations. It is not expected to be exact in realistic applications because the underlying assumption, such as neglecting transverse mixing altogether, may not hold. We quantify the error introduced by the approximation for the hypothetical case of a two-dimensional, binary aquifer made of highly-permeable, non-reactive and low-permeable, reactive materials releasing dissolved organic matter acting as electron donor for aerobic respiration and denitrification. The kinetically controlled reactions are catalyzed by two non-competitive bacteria populations, enabling microbial growth. Even though the initial biomass concentrations were uniform, the interplay between transport, non-uniform electron-donor supply, and bio-reactions led to distinct spatial patterns of the two types of biomass at late times. Results obtained by mapping the exposure-time based results to the two-dimensional domain are compared with simulations based on the two-dimensional, spatially explicit advection-dispersion-reaction equation. Once quasi-steady state has been reached, we find a good agreement in terms of the chemical-compound concentrations between the two approaches inside the reactive zones, whereas the exposure-time based model is not able to capture reactions occurring in the zones with zero electron-donor release. We conclude that exposure-time models provide good approximations of nonlinear bio-reactive transport when transverse mixing is not the overall controlling process and all reactions are essentially restricted to distinct reactive zones.

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1. Introduction

Significant spatial variability of aquifer properties determines the fate of reactive components in groundwater. While the heterogeneity of physical properties (mainly hydraulic conductivity) determines the spatial structure of the groundwater flow field, the spatial variability of geochemical properties establishes different aquifer reactivities within the domain. Scheibe et al. (2006) recommended that the effects of coupled physical and biogeochemical heterogeneities should be jointly considered in reactive transport and bioremediation modeling. As tested by Cunningham and Fadel (2007) and Atchley et al. (2014), among others, both positive and negative correlations between physical and geochemical heterogeneities substantially modify the propagation and fingering of reactive fronts in porous media. The interplay between spatial variability of linear sorption (both in local equilibrium and kinetic) and hydraulic conductivity has been the subject of intensive stochastic analysis (e.g., Burr et al., 1994; Espinoza and Valocchi, 1997; Cushman et al., 1995; Cvetkovic et al., 1998). Dentz et al. (2011) analyzed kinetic dissolution-precipitation reactions in physically and chemically heterogeneous domains, highlighting that physical and chemical heterogeneity cannot be upscaled separately. Fully analytical treatment of joint physico-chemical heterogeneity in transport with nonlinear reactions, however, is inherently difficult, so that numerical simulations of such systems appear indispensable.

Most natural redox reactions in groundwater are catalyzed by microbes, which depend on particular chemical environments but also change them. Ginn et al. (2002) highlighted the need for new modeling approaches accounting for the time that a microorganism has been exposed to nutrients, the dynamics of biomass growth under changing chemical conditions, and the effects of heterogeneity on microbial populations. Likewise, Mohamed et al. (2010) observed that the mass discharge of contaminants undergoing microbial degradation is highly sensitive to chemical heterogeneity because the availability of substrates controls microbial activity. Tufenkji (2007) stressed the need of understanding the influence of physical and geochemical heterogeneity on microbial transport in porous media.

A key challenge for numerical bioreactive transport modeling in heterogeneous media is to allocate restricted computational resources: Uncertainty of the spatial distribution of aquifer properties requires multiple simulations with different parameter combinations (in the simplest version as Monte-Carlo simulations) but the computational effort for a single simulation run using multi-dimensional spatially explicit methods may be so high that it restricts the number of possible runs. In certain environmental settings, travel-time based bioreactive transport models are a feasible alternative to the traditional spatially-explicit advective-dispersive-reactive models due to the simplification of a multi-dimensional problem to a quasi one-dimensional one (see Sanz-Prat et al., 2015, 2016). The travel time $\tau(\mathbf{x})$ [T] is defined as the time that a particle needed to travel from the inlet boundary to the observation point with spatial coordinates \mathbf{x} [L]. Due to dispersion, the travel time at a point is a probability density function, $p(\tau(\mathbf{x}))$, rather than a unique value, but its mean value (also denoted the mean groundwater age) and spread can

easily be computed by temporal-moment generating equations (Harvey and Gorelick, 1995; Goode, 1996). The main assumption of travel-time based reactive-transport models is that reactive fronts are aligned with groundwater isochrones. This assumption holds under idealized conditions without groundwater mixing, that is, without dispersion. Also flow should be at steady state, the distribution of the reactants should be uniform across the entire inlet boundary, and the biogeochemical parameters must be uniform within the domain.

Various analyses have been based on the assumption of strictly advective transport, facilitating a Lagrangian analysis of solute transport in multi-dimensional domains (e.g., Simmons, 1982; Dagan and Nguyen, 1989; Shapiro and Cvetkovic, 1988; Dagan et al., 1992; Cvetkovic et al., 1992). Conceptually identical to the kinematic-Lagrangian framework is the assumption of transport occurring by advection only in independent stream tubes that do not interact with each other (e.g., Crane and Blunt, 1999). This framework is well suited to address reactive transport, at least in steady-state flow or if there are no reactants in an immobile phase (e.g., Dagan and Cvetkovic, 1996; Cvetkovic and Dagan, 1996; Kaluarachchi et al., 2000; Simmons et al., 1995; Diem et al., 2013). Ginn et al. (1995) simulated coupled microbial growth and non-linear reactive transport of a single solute in one-dimensional stream tubes and addressed the effects of heterogeneity by considering a distribution of velocities among individual stream tubes, meeting the travel-time distribution in the outlet of the original multi-dimensional domain. This method enables the upscaling of microbially influenced transport in heterogeneous domains without relying on Fickian macrodispersion, which has been shown to introduce artifacts in bioreactive transport already by Molz and Widdowson (1988). Stream-tube formulations considerably reduce the computational effort making Monte-Carlo simulation to analyze the effects of spatially variable physical properties on reactive transport feasible. Cirpka and Kitanidis (2000a) denoted the latter method the advective-dispersive stream-tube approach and suggested parameterizing the dispersivities of the independent stream tubes by analyzing the width of point-like measured breakthrough curves of conservative tracers (Cirpka and Kitanidis, 2000b). Ginn (2001) and Ginn et al. (2001) extended the approach by allowing longitudinal dispersive mixing within the stream tubes. Cirpka (2002) used effective longitudinal dispersion coefficients derived by linear stochastic theory (Dentz et al., 2000; Fiori and Dagan, 2000) for the within-stream-tube mixing, and the difference between ensemble and effective dispersion to parameterize the apparent velocity distribution among the independent stream tubes. In all these studies, the intrinsic rate coefficients and the initial biomass distribution were assumed to be spatially uniform. Microbial biomass, and thus the concentration of the reaction catalyst, was allowed to vary but it organized itself due to the interplay of advective (or advection-dominated) transport, reactions, and microbial dynamics (growth, decay, in some cases transport of microbes). In two preceding studies, we have analyzed the validity of travel-time based models for bioreactive transport subject to small, but non-zero transverse mixing (Sanz-Prat et al., 2015) and under the additional influence of time-period fluctuations of the magnitude of flow (Sanz-Prat et al., 2016). As a reactive

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