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A dual flowing continuum approach to model denitrification experiments in porous media colonized by biofilms

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ABSTRACT

We present a modeling exercise of solute transport and biodegradation in a coarse porous medium widely colonized by a biofilm phase. Tracer tests in large laboratory columns using both conservative (fluorescein) and biodegradable (nitrate) solutes are simulated by means of a dual flowing continuum approach. The latter clearly distinguishes concentrations in a flowing porous phase from concentrations conveyed in the biofilm. With this conceptual setting, it becomes possible to simulate the sharp front of concentrations at early times and the flat tail of low concentrations at late times observed on the experimental breakthrough curves. Thanks to the separation of flow in two phases at different velocities, dispersion coefficients in both flowing phases keep reasonable values with some physical meaning. This is not the case with simpler models based on a single continuum (eventually concealing dead-ends), for which inferred dispersivity may reach the unphysical value of twice the size of the columns. We also show that the behavior of the dual flowing continuum is mainly controlled by the relative fractions of flow passing in each phase and the rate of mass transfer between phases. These parameters also condition the efficiency of nitrate degradation, the degradation rate in a well-seeded medium being a weakly sensitive parameter. Even though the concept of dual flowing continuum appears promising for simulating transport in complex porous media, its inversion onto experimental data really benefits from attempts with simpler models providing a rough pre-evaluation of parameters such as porosity and mean fluid velocity in the system.

1. Introduction

The activity of bacteria is now widely exploited in groundwater treatments for either ex-situ drinkable water production or in-situ aquifer decontamination. The bacterial growth onto the solid phase of a porous/fractured medium is stimulated by the presence and/or the addition of an appropriate feeding substrate. Rittmann (1993) suggested that this growth was preferentially initiated in the form of micro-colonies spread in an oligotrophic environment and then evolving toward the formation of a continuous biofilm when the load in substrate increased. The biofilm is therefore made of bacterial micro-colonies embedded in a matrix of Extra Cellular Polymers (ECP) riddled of interstitial open voids and connected channels (see, e.g., Characklis, 1989; Lewandowski, 2000). Because the biofilm is the place of reaction occurrences, it becomes a key for success of biological treatments in dynamic systems, the solute degradation depending on transport in the porous medium and also on mass transfers from the fluid phase to the bacterial cells. The conception, the functioning of these biological technologies and their expected efficiency are to some extent grounded in modeling the coupling between transport and mass degradation in the presence of a biofilm.

Modeling reactive transport of a biodegradable solute is a multi-faceted and multi-scale problem with a reaction occurring at the scale of the bacteria but also needing for integration

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at the operational scale of a few meters or so. Solving the problem at the metric scale should ideally encompass all the local mechanisms and processes acting at the microscopic and pore scales. In the continuous macroscopic models, the effects of microscopic features are substituted by effective mechanisms and their associated parameters at the Darcy's scale by following two main methods: 1— up-scaling techniques seeking the physical meaning and the mathematical formalism of the effective mechanisms and parameters at the large scale; 2— the so-called heuristic approach conjecturing a priori on the mechanisms at the large scale and then seeking their parameters by facing model results to data (i.e., the general framework of inversion exercises).

On the side of rigorous up-scaling exercises, the works of Wood and Whitaker (1998) and Wood et al. (2002) developed the passage from the single bacteria to the whole biofilm taken as a continuous phase by means of volume averaging techniques (Whitaker, 1999). The most recent works along this line tackle with the next step of passing from the scale of the biofilm to the operational metric — decametric scale relevant to simulate and foresee the behavior of water treatment systems (Aspa et al., 2011; Golfier et al., 2009; Orgogozo et al., 2010). The up-scaling procedures rely on the calculation of effective velocities, diffusion and dispersion tensors while accounting for the geometry and the volumetric fractions of the different phases in the medium, e.g., bacterial cells versus ECP matrix, biofilm versus fluid phase. It must be acknowledged however that the three-dimensional imaging and the geometric characterization of a biofilm phase colonizing a porous medium are challenging because of some features of the object, for instance: the biofilm opacity, the multiple porosities, the multiple compounds, etc. Investigation techniques especially set-up for imaging biofilms are still in their phase of development (confocal microscopy, X-ray tomography, magnetic resonance microscopy) and the same comment also applies to the numerical processing of images resulting from these techniques (Beyenal and Lewandowski, 2000; Davit et al., 2011; Iltis et al., 2011; Seymour et al., 2007).

The current experimental difficulties mentioned above are conducive to: 1— the approximation of the macroscopic transport parameters by means of analytical or numerical solutions based on synthetic geometrical simplifications of the biofilm phase; 2— the simulation of the biofilm morphology and the subsequent properties by means of bacterial growth modeling (Kreft et al., 2001; Picioreanu et al., 1999a, 1999b). Despite these techniques may allow bridging a gap between a homogenized model and the micro-scale physics, the influence of the simplifications on effective parameters is not clear and motivates ongoing studies (Orgogozo et al., 2010; Aspa et al., 2011). Incidentally, the heavy numerical efforts invested in the passage from bacterial cell to biofilm do not bring confidence in our current capabilities of up-scaling transport with the same techniques at the metric–decametric scale.

The heuristic approach deliberately overlooks the structural and morphological heterogeneity of the biofilm and states the problem directly at the macroscopic scale by conjecturing on the prevailing mechanisms and the associated mathematical formalism. This method seems the best suited to operational applications in view of the current computation capabilities and of the few data available in general on reactive transport at laboratory and field scales. Two main groups of heuristic models can be distinguished according to the way the solute is conveyed from the fluid phase to the biofilm.

The first group merges the porous medium and the biofilm into a single-continuum subjected to advection, dispersion and local equilibrium of the concentrations between the flowing fluid and the biofilm. Stated otherwise, this type of model overlooks the eventual effects of diffusion added by the biofilm, or more precisely, considers that the diffusion time in the biofilm is negligible compared with the advection–dispersion time in the fluid phase (Baveye and Valocchi, 1989; Borden, 2007; Killingstad et al., 2002). In the case of well-seeded media with a non-negligible volumetric fraction of biofilm, the single-continuum model results in the inference of excessive dispersion coefficients as the consequence of solute travel times spanning wide ranges (Hill and Sleep, 2002; Sharp et al., 1999; Sharp et al., 2005).

By considering that the growth of a biofilm in a porous medium yields a dual-porosity system, i.e., a main porosity of “channels” and a secondary porosity including dead-ends and micro-pores in the biofilm, the second group of models assumes the existence of mass transfers between a mobile fluid phase and an immobile biofilm phase. This transfer is sometimes written as a diffusion equation with approximations on surface area and depth of biofilm involved in the mechanism (Molz et al., 1986; Taylor et al., 1990; Widdowson et al., 1988). Most often, the mass transfer is simply ruled by a first-order kinetics without any prior guess on the structure of the interface between the fluid and the biofilm (Baveye and Valocchi, 1989; Wood et al., 1994). The results reported by Taylor and Jaffé (1990), Seifert and Engesgaard (2007), show that mass transfers cannot alone explain the flat-tailed distributions of long resting times observed on numerous experimental breakthrough curves. In the end, and even though heuristic models are promising tools for operational applications, their conceptualization and their fundaments miss some specific features of porous media well-seeded in bacteria. One can also raise that most studies on heuristic models including a biofilm phase put emphasis on the identification of transport parameters but generally overlook the reaction part.

The goal of the present work is to re-handle and enrich a heuristic approach to transfer in a porous medium densely colonized by a biofilm. We assume that both compartments, fluid and biofilm, are flowing media linked by a kinetically controlled mass transfer. By construction, this type of model can mimic large solute dispersion by simply separating the travel time distribution in two modes, i.e., the first-one associated with the fluid velocity in the porous medium and the second-one associated with the velocity in the biofilm. A direct consequence is that the dispersion coefficients attached to each flowing continuum keep reasonable values with some physical meaning. We show also that the dual flowing continuum can be reduced to simpler models either to retrieve results from previous studies, or more interestingly, to proceed by steps and facilitate inversion of experimental data (see hereafter).

Regarding experimental data, tracer tests in large laboratory columns performed according to three different settings are available: 1— an inert solute transported in a non-colonized porous medium, 2— an inert solute in the same porous medium densely colonized by a biofilm, 3— a biodegradable solute