Journal of Contaminant Hydrology 191 (2016) 1–18

Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

journal homepage: www.elsevier.com/locate/jconhyd

Research paper

Global sensitivity analysis and Bayesian parameter inference for solute transport in porous media colonized by biofilms

A. Younes [a,b,*], F. Delay [a], N. Fajraoui [a], M. Fahs [a], T.A. Mara [c]

[a] LHyGES, Université de Strasbourg/EOST, CNRS, 1 rue Blessig, 67084 Strasbourg, France
[b] IRD UMR LISAH, F-92761 Montpellier, France
[c] Université de La Réunion, PIMENT, 15 Avenue René Cassin, BP 7151, 97715 Moufia, La Réunion, France

ARTICLE INFO

Article history:
Received 23 November 2015
Received in revised form 22 March 2016
Accepted 30 April 2016
Available online 3 May 2016

Keywords:
Reactive transport
Dual flowing continuum
Biofilms
Bayesian inference
Global sensitivity analysis
Sparse polynomial chaos expansion

ABSTRACT

The concept of dual flowing continuum is a promising approach for modeling solute transport in porous media that includes biofilm phases. The highly dispersed transit time distributions often generated by these media are taken into consideration by simply stipulating that advection–dispersion transport occurs through both the porous and the biofilm phases. Both phases are coupled but assigned with contrasting hydrodynamic properties. However, the dual flowing continuum suffers from intrinsic equifinality in the sense that the outlet solute concentration can be the result of several parameter sets of the two flowing phases. To assess the applicability of the dual flowing continuum, we investigate how the model behaves with respect to its parameters. For the purpose of this study, a Global Sensitivity Analysis (GSA) and a Statistical Calibration (SC) of model parameters are performed for two transport scenarios that differ by the strength of interaction between the flowing phases.

The GSA is shown to be a valuable tool to understand how the complex system behaves. The results indicate that the rate of mass transfer between the two phases is a key parameter of the model behavior and influences the identifiability of the other parameters. For weak mass exchanges, the output concentration is mainly controlled by the velocity in the porous medium and by the porosity of both flowing phases. In the case of large mass exchanges, the kinetics of this exchange also controls the output concentration.

The SC results show that transport with large mass exchange between the flowing phases is more likely affected by equifinality than transport with weak exchange. The SC also indicates that weakly sensitive parameters, such as the dispersion in each phase, can be accurately identified. Removing them from calibration procedures is not recommended because it might result in biased estimations of the highly sensitive parameters.

1. Introduction

Flow and reactive transport in porous media colonized by biofilms are of particular interest in groundwater treatments and in many environmental engineering systems. In this context, several modeling approaches have been developed at the Darcy scale, such as up-scaling techniques or heuristic approaches including flow and transport either in a single continuum, in a single flowing continuum plus a stagnant phase, or in a dual flowing continuum.

Usually, the heuristic single continuum model only accounts for advection–dispersion in the porous medium. In the case of well-seeded media with a non-negligible volumetric fraction of biofilm, the single continuum is prone to the inference of unphysical excessive dispersion coefficients (Sharp et al., 1999; Hill and Sleep, 2002; Sharp et al., 2005). This problem is overcome when the colonized porous medium

* Corresponding author at: IRD UMR LISAH, F-92761 Montpellier, France.
E-mail address: younes@unistra.fr (A. Younes).

http://dx.doi.org/10.1016/j.jconhyd.2016.04.007
0169-7722/

is modeled as the sum of a flowing porous phase and a stagnant biofilm phase (Baveye and Valocchi, 1989; Clement et al., 1997; Phanikumar et al., 2005; Seifert and Engesgaard, 2007). When solute mass transfer between the flowing and the stagnant phases is ruled by a first-order kinetic law, the approach is equivalent to the so-called dead-end model where the system is formed by a main porosity of flowing channels and a secondary porosity of non-flowing dead-ends (Coats and Smith, 1964; De Smedt and Wierenga, 1979; Van Genuchten and Wierenga, 1976).

A dual flowing continuum approach was proposed by Delay et al. (2013) to simulate laboratory experiments of mass transfer in a porous medium colonized by a biofilm. The concept was shown to be appropriate in the case of widely colonized media, as the single continuum approach led to unphysical results with inferred dispersivity values larger than the size of the modeled system (Delay et al., 2013). The main characteristic of the dual flowing continuum is to convey the solute by advection–dispersion in both the porous and the biofilm phases, each phase being assigned with specific transport properties. Interaction between the two continua is maintained by solute mass exchange with first-order kinetics. The assumption of non-negligible flow in the biofilm phase is supported by several works (De Beer et al., 1997; Stoodley et al., 2008, among others) and the following observations. At the sample scale, the biofilm phase with its skeleton of extra cellular polymers is rarely a simple coating of the grains in a porous medium. The biofilm is usually a phase of porous dendritic filaments opened enough to convey flow. This structure was evidenced by X-ray tomography in many samples of 1–10 cm size (e.g., Davit et al., 2011, Iltis et al., 2011). At a larger scale, for example, in a laboratory column or in an in-situ pilot study of decontamination, colonized porous media often appear highly dispersive due to contrasted fluid velocities in partly separated flow paths of the medium (e.g., Roubinet et al., 2013). The dual flowing continuum approach clearly demonstrates the capability to reproduce a large apparent solute dispersion while keeping local dispersion coefficients to small values. This feature was first raised by the pioneer work of Gerke and Van Genuchten (1993) introducing a dual porosity model with two flowing phases (as is the dual flowing continuum) to model flow and transport in fractured porous media with non-negligible flow in the matrix hosting the fracture network (see also Hutson and Wagenet, 1995; Gwo et al., 1996, Fahs et al., 2013). Notably, the dual flowing continuum approach is versatile because it can be degraded into a single flowing continuum or a single continuum plus a stagnant phase.

The dual flowing continuum model introduces however several parameters, such as transport parameters in each phase and exchange rate between the porous medium and the biofilm (see Section 2). In practical applications, e.g., designing a water decontamination plant that utilizes bacteria for the natural eradication of various contaminants, most of the model parameters cannot be measured directly and should be estimated using an inversion exercise. In the case of a dual flowing continuum, the inversion can be challenging due to the non-identifiability of the parameters. By duplicating the number of parameters (as the consequence of increasing the number of transport equations), the dual flowing continuum increases the risk of introducing irrelevant parameters to which the model response is not notably sensitive. Moreover, calculations using two transport equations of the same type for both the porous and the biofilm phases may yield to model equifinality, implying that different parameter combinations between two transport equations can fit the model responses to data.

Hence, the first objective of this work is to investigate how the dual flowing continuum model behaves with respect to its parameters. To this aim, we analyze the impact of the parameters on the output concentration via a Global Sensitivity Analysis (GSA). The GSA is useful to: (*i*) assess the applicability of a model; (*ii*) distinguish between influential (that contribute the most to the variability of model outputs) and non-influential parameters; and finally, (*iii*) understand the behavior of the modeled system.

The second objective is to assess the identifiability of the different parameters from a set of noisy data using Statistical Calibration (SC). In this work, the SC is performed with the $\text{DREAM}_{(\text{ZS})}$ software (Laloy and Vrugt, 2012) based on the Markov Chain Monte Carlo process (MCMC). The MCMC inversion provides not only the best estimates of the parameters but allows for exploring the entire parameter space of the posterior distribution of the parameters. MCMC also provides the pairwise parameter correlations and the uncertainty on model predictions.

Furthermore, according to Delay et al. (2013), the kinetic rate of mass transfer between the two flowing continua deeply impacts the shape of the solute breakthrough curves which in turn can modify the model behavior and the identifiability of the model parameters. To investigate this effect, two contrasted flowing scenarios are considered in this study. These scenarios manipulate synthetic data drawn from numerical simulations to avoid the addition of uncontrolled noise from actual experiments that could bias conclusions. However, these scenarios are based on actual experiments (reported in Delay et al., 2013) performed in large laboratory columns and developed to address the feasibility of a water decontamination (denitrification) plant in the field. Both scenarios deal with the injection of a conservative tracer in a porous medium colonized by a stationary biofilm phase. The bacteria are assumed immobile with limited bacterial concentrations moved by the fluid. Bacterial growth or decay is not considered to both simplify the problem and mimic usual forcing conditions in aquifers such as weakly transient flow at small velocities (few shear stress acting on bacteria), stable physico-chemical conditions and enough dissolved organic substrate for feeding the bacterial mass. The scenarios differ mainly by the (simulated) maturation degree of the biofilm phase in the porous medium. The first scenario addresses a matured biofilm encysted in the porous medium by a thick skeleton of extra cellular polymers, which is not favorable for the solute mass exchange between the porous phase and the biofilm. Alternatively, the second scenario handles an immature biofilm, which provides a high exchange rate with the porous medium.

The structure of the present study is as follows. Section 2 presents the dual flowing continuum approach to solute transport. Section 3 reports on the transport scenarios and the techniques used to perform GSA and SC for these scenarios. Then, Section 4 discusses the GSA and SC results. Emphasis is