Journal of Hydrology 541 (2016) 1020–1029

Contents lists available at ScienceDirect

Journal of Hydrology

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

Research papers

A multi-scale Lattice Boltzmann model for simulating solute transport in 3D X-ray micro-tomography images of aggregated porous materials

Xiaoxian Zhang [a,*], John W. Crawford [a], Richard J. Flavel [b], Iain M. Young [c]

[a] Department of Sustainable Soils and Grassland Systems, Rothamsted Research, West Common, Harpenden, Hertfordshire AL5 2JQ, UK
[b] School of Environment & Rural Science, The University of New England, Armidale, SW 2351, Australia
[c] School of Life and Environmental Sciences, Faculty of Science, The University of Sydney, NSW 2006, Australia

A R T I C L E   I N F O

Article history:
Received 3 June 2016
Received in revised form 1 August 2016
Accepted 8 August 2016
Available online 9 August 2016
This manuscript was handled by P. Kitanidis, Editor-in-Chief, with the assistance of Juan V. Giraldez, Associate Editor

Keywords:
X-ray images
Grey Lattice Boltzmann model
Solute transport
Pore-scale modelling
Multi-scale model
Soil

A B S T R A C T

The Lattice Boltzmann (LB) model and X-ray computed tomography (CT) have been increasingly used in combination over the past decade to simulate water flow and chemical transport at pore scale in porous materials. Because of its limitation in resolution and the hierarchical structure of most natural soils, the X-ray CT tomography can only identify pores that are greater than its resolution and treats other pores as solid. As a result, the so-called solid phase in X-ray images may in reality be a grey phase, containing substantial connected pores capable of conducing fluids and solute. Although modified LB models have been developed to simulate fluid flow in such media, models for solute transport are relatively limited. In this paper, we propose a LB model for simulating solute transport in binary soil images containing permeable solid phase. The model is based on the single-relaxation time approach and uses a modified partial bounce-back method to describe the resistance caused by the permeable solid phase to chemical transport. We derive the relationship between the diffusion coefficient and the parameter introduced in the partial bounce-back method, and test the model against analytical solution for movement of a pulse of tracer. We also validate it against classical finite volume method for solute diffusion in a simple 2D image, and then apply the model to a soil image acquired using X-ray tomography at resolution of 30 μm in attempts to analyse how the ability of the solid phase to diffuse solute at micron-scale affects the behaviour of the solute at macro-scale after a volumetric average. Based on the simulated results, we discuss briefly the danger in interpreting experimental results using the continuum model without fully understanding the pore-scale processes, as well as the potential of using pore-scale modelling and tomography to help improve the continuum models.

1. Introduction

Numerical modelling of water flow and solute transport in soils plays an important role in many areas ranging from design of remediating strategies for contaminated soils and aquifers to calculating uptake of water and solutes by plant roots. While practical modelling relies on continuum approaches using a volumetric average to phase out the detailed pore geometries, it is the invisible processes occurring at pore scale of micron(s) that underpin the phenomena measurable at macroscopic scales (O'Donnell et al., 2007). Understanding the pore-scale processes is hence essential to improving continuum models and has received increased interest since the 1950s (Blunt et al., 2013; Fatt, 1956; Reeves and Celia, 1996; Zhang et al., 2008). Because of the opaque nature of soils, however, the early pore-scale models were mainly based on idealizations by approximating the complicated pore geometry in soils with a regular lattice network encompassing spherical pore bodies linked by cylindrical pore throats with each pore or throat occupied by one fluid (Blunt, 2001; Celia et al., 1995; Fatt, 1956).

The advent of tomography technologies over the past decade, particularly the X-ray micro-computed tomography (μCT), neutron tomography and focused ion beam/scanning electron microscopy (FIB/SEM), has made non-intrusively visualizing the interior structures of opaque materials at a resolution of a few nanometres feasible (Oburger and Schmidt, 2016; Wildenschild et al., 2002; Zhang et al., 2014). This, together with the development in computational fluid dynamics, especially the Lattice Boltzmann (LB) method (Chen and Doolen, 1998) and the smooth particle hydrodynamics (Tartakovsky et al., 2007), has created a renewed interest in pore-scale modelling in a wide range of areas including fuel cells and carbon sequestration (Chen et al., 2009; Gao et al., 2012;

* Corresponding author.
E-mail address: xiaoxian.zhang@rothamsted.ac.uk (X. Zhang).

http://dx.doi.org/10.1016/j.jhydrol.2016.08.013
0022-1694/

Huang et al., 2011; Pot et al., 2015). Over the past two decades, tremendous progresses have been made in developing LB models for simulating water flow and solute transport at pore scale in porous materials (Ginzburg, 2005; Kang et al., 2007; Knutson et al., 2005; Zhang et al., 2008; Zhang and Lv, 2007). However, most of these models were based on binary images assuming that fluids and chemicals can only move in the void phase and the perceived solid phase as impermeable.

Most soils are naturally hierarchical with their pore sizes ranging from less than one micron (for inter-aggregates pores) to millimetres (for intra-aggregate pores). In imaging such soils using μCT, one can only identify the pores which are greater than the image resolution, while treats smaller-scale pores as solid (Luo et al., 2010; Ma et al., 2015; Peng et al., 2014). Thus, the so-called solid in the X-ray images is in all probability a 'grey' phase, perhaps containing substantial connected pores capable of conducting fluids and solute. How to deal with such images has been an interest in pore-scale modelling, and modified LB models have been developed to simulate fluid flow in them. One approach is simply to add an extra force to the standard LB model to describe the resistance caused by the solid matrix in the grey phase to fluid flow (Kang et al., 2002). Another approach is to modify the bounce-back method by partly bouncing back particles to describe the resistance of the solid matrix in the grey phase to fluid movement; the portion of the particles being bounced back is related to the permeability of the grey phase (Walsh et al., 2009; Zhu and Ma, 2013). A comparison of different approaches available in the literature for simulating fluid flow in images consisting of grey phase was given in a recent review by Ginzburg (2016).

In contrast to fluid flow, research on solute transport in images acquired using tomography is limited. The available LB models for solute transport include the single relaxation-time (BGK) model (Zhang et al., 2002a, 2002b), the multiple relaxation-time (MRT) model (Yoshida and Nagaoka, 2010) and the two relaxation-times model (TRT) (Ginzburg, 2007; Vikhansky and Ginzburg, 2014). In simulating chemical transport in heterogeneous media where the diffusion coefficient is spatially variable, the BGK model solves it using a spatially variable relaxation-time parameter (Sukop and Cunningham, 2014; Zhang et al., 2002b). In contrast, the MRT and TRT model could solve this heterogeneity by tuning some of its free parameters (Ginzburg, 2007; Ginzburg and d'Humieres, 2007; Ginzburg et al., 2010).

In the past, we have developed a BGK model to simulate solute transport in unsaturated soils where the dispersion coefficient varies with water content (Zhang et al., 2002b). It is worth pointing out that these models require the dispersion coefficient to be spatially continuous. For solute transport in X-ray images encompassing pores and permeable grey phase, its diffusion coefficient has an abrupt change across the pore-solid interface. When applying the BGK models to such images, the discontinuity of the diffusion coefficient at the pore-solid interface could give rise to significant errors. To overcome this problem, we propose a modified LB model in this paper based on the partial bounce-back approach to simulate chemical transport in 3D soil images acquired using tomography. The method naturally reduces to the standard bounce-back method when the grey phase becomes impermeable and to the standard streaming step for solute in the void space. We derive the relationship between the parameter introduced in the partial bounce-back method and the diffusion coefficient for solute moving in both the void space and the grey phase, and demonstrate its improvement in comparison with the available BGK models. We then apply the model to a soil image acquired using X-ray tomography at resolution of 30 μm in an attempt to elucidate how the ability of the grey phase at micro-scale to diffuse chemicals could affect the average solute transport behaviour at macro-scale after a volumetric average. In particular, we analyse the danger in interpreting experimental data using the continuum approaches without fully understanding the pore-scale processes, and the potential of using the tomography and pore-scale models to help improve the macroscopic continuum models.

2. The model

Both water flow and chemical transport were simulated using the LB models. The model for water flow was based on the standard single-relaxation approach and its detailed description is available in the literature (Ginzburg, 2016; Zhang et al., 2005, 2008; Zhu and Ma, 2013). Here, we focus on chemical transport assuming that the detailed water velocity at pore scale had already been obtained. Like other LB models, the proposed LB model consists of a collision step and a streaming step in each time step. The collision takes place at time t and is to calculate

$$f_i^c(x,t) = f_i(x,t) - \tau\left[f_i(x,t) - f_i^{eq}(x,t)\right], \tag{1}$$

where $f_i(x,t)$ is the particle distribution function – the probability of finding a particle at location x and time t moving with velocity ζ_i, $f_i^{eq}(x,t)$ is the equilibrium distribution function, and τ is a relaxation-time parameter controlling the rate of $f_i(x,t)$ approaching $f_i^{eq}(x,t)$.

The streaming step is to move the collision result calculated from Eq. (1) to location $x + \zeta_i\delta t$ at the end of each time step, δt. During the streaming step, if there is no solid barrier between x and $x + \zeta_i\delta t$, the streaming moves the particle distribution function at the end of each time step to become $f_i(x + \zeta_i\delta t, t + \delta t) = f_i^c(x,t)$. In contrast, if impermeable solid is present within $(x + \zeta_i\delta t/2, x + \zeta_i\delta t)$ but absent in $(x, x + \zeta_i\delta t/2)$, the bounce-back method is often used to treat the impermeable solid by bouncing the in-coming particle towards the solid back to where the particle emanates at the beginning of the time step, i.e., $f_i(x + \zeta_i\delta t, t + \delta t) = f_{\hat{i}}^c(x + \zeta_i\delta t, t)$, where the subscript $\hat{i}$ represents the opposite direction of i, i.e., $\zeta_{\hat{i}} = -\zeta_i$.

For chemical transport in images acquired using tomography where the solid (i.e., the grey phase) is in reality partly permeable but deemed solid due to the aforementioned restrictions in μCT, we propose the following partial bounce-back method to describe the resistance caused by the permeable solid phase to chemical transport:

$$f_i(x_o + \zeta_i\delta t, t + \delta t) = n_s f_{\hat{i}}^c(x_o + \zeta_i\delta t, t) + (1 - n_s) f_i^c(x_o, t), \tag{2}$$

where $0 \leqslant n_s \leqslant 1$ is a parameter with $n_s = 0$ representing the chemical in the pores, and $0 < n_s$ representing the chemical in the grey phase. When $n_s = 1$, the grey phase becomes impermeable and Eq. (2) reduces to the standard bounce-back method.

The interest in solute transport is concentration. We hence follow Zhang et al. (2008) assuming that the particle distribution functions move in seven directions with velocities $\zeta_0 = (0,0,0)$, $\zeta_{1,3} = \pm(\delta x/\delta t, 0, 0)$, $\zeta_{2,4} = \pm(0, \delta x/\delta t, 0)$ and $\zeta_{5,6} = \pm(0, 0, \delta x/\delta t)$, where δt and δx are time step and the side length of the 3D voxels respectively. For ease of analysis, in what follows we will normalize the space by δx and the time by δt. The equilibrium distribution function associated with each velocity is defined as follows:

$$\begin{aligned}
&f_i^{eq} = c_i(\varepsilon + 3.5U_\alpha e_{i\alpha})/7, \\
&U_\alpha = \frac{\tau u_\alpha}{\tau - 2n_s[(1 - 0.5\tau) + n_s(\tau - 1)]}, \quad n_s < 1, \\
&U_\alpha = 0, \quad n_s = 1,
\end{aligned} \tag{3}$$

where ε is the porosity of the grey phase ($\varepsilon = 1$ for the void space, and $\varepsilon = 0$ when the solid phase becomes impermeable), u_α is the bulk water velocity component in the x_α direction (equivalent to the *Darcy* flow rate in the grey phase), and $\boldsymbol{e}_i = \zeta_i\delta t/\delta x$. The mass