



# Impact of saturation on mass transfer rate between mobile and immobile waters in solute transport within aggregated soils



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## SUMMARY

Solute transport in aggregated soils is controlled by pores both inside and between the aggregates. Because the intra-aggregate pores are much smaller than the inter-aggregate pores, in chemical transport modelling the water in the former was often assumed to be immobile in comparison with water in the latter. How to describe mass transfer between the two waters has been studied intensively for saturated soils but poorly for unsaturated soils. In this paper, we investigated this using pore-scale modelling and tomography. The binary structures of porous materials acquired using tomography in our previous work served as the aggregated soils. Since the sizes of the intra-aggregate pores were smaller than the resolution of the tomography, they cannot be explicitly resolved in the tomography. As a result, the solids in the binary structures were porous aggregates and their impact on solute movement was described by an effective diffusion coefficient. In all simulations, the aggregates were assumed to be fully saturated and water distribution between the aggregates was determined by inter-aggregate pore sizes and pore connectedness. Solute movement from water within the inter-aggregates into the aggregates under different saturations was simulated using a pore-scale model. The simulated concentration and flux at pore scale were spatially averaged, and they were then used to calculate the volumetric average mass transfer rate between the two waters. The calculated average mass transfer rates were linked to the memory function widely used in the literature to model solute transport in structured soils. The results indicate that the commonly-used linear mobile–immobile transfer model with its transfer rate coefficient proportional to water content cannot fit the memory function calculated at any saturation. We fitted the simulated results to an empirical formula. The comparisons reveal that in the earlier stage, the memory function decreases with time in a power-law, and in the later stage it decreases exponentially. The time it takes such a transition to complete increases as the saturation decreases. We also explained and proved how to extend these results to simulate reactive solutes.

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## 1. Introduction

Solute transport and reaction in aggregated soils plays an important role in many areas including nutrient uptake by plant roots and pollutant leaching in soils. In chemical transport modelling, because the pores inside the aggregates are much smaller than the pores between the aggregates, the water in the former was often assumed to be immobile in comparison with water in the latter (Knabner et al., 1996; Yang et al., 2014). If there is a concentration difference between the two waters, the chemical will move from one to another, driven by molecular diffusion. How to describe such a mass transfer is an important issue in

chemical transport modelling (Haggerty and Gorelick, 1995; Simunek et al., 2003).

An earlier approach to describe mass transfer in structured media is the mobile–immobile model proposed by Coats and Smith (1964), in which the mass transfer rate was assumed to be proportional to concentration difference between the two waters. Theoretical analysis later proved that the underlying assumption of this model is that the medium is a packing of isolated spherical aggregates and solute transport in the spheres is independent of each other (Haggerty and Gorelick, 1995). This is an obvious oversimplification, and its application to real soils often requires the mass transfer rate to increase with pore-water velocity in order to match the experimental data (Bromly and Hinz, 2004; Gao et al., 2009; Pang et al., 2002). Over the past two decades, tremendous efforts have been expended in attempts to improve the

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description of such mass transfers in structured media; a number of approaches with varying difficulties have been proposed, including the multi-rate adsorptive model (Russo et al., 2010), fractal model (Schumer et al., 2003) and continuous time random walk model (Berkowitz et al., 2006). Recently, Cvetkovic (2012) also presented a general model to describe the mass exchange for modelling chemical transport in fractured media.

Most available research on mass transfer in structured media focused on saturated conditions (Zoia et al., 2010). For unsaturated aggregated soils, van Genuchten and Wierenga (1976) proposed to extend the mobile-immobile model of Coats and Smith by modifying the mass transfer rate to be proportional to the saturation of the inter-aggregate pores. This appears to be the main model currently in use in the literature to simulate chemical transport in unsaturated soils (Gardenas et al., 2006; Gerke and van Genuchten, 1993; Simunek et al., 2003; van Genuchten and Wierenga, 1976). Given that this linear mobile-immobile model had been found inadequate for some saturated structured media (Cvetkovic, 2012), its application to unsaturated soils could give rise to errors under certain circumstances as the mass transfer in unsaturated aggregated soils is more complicated, depending not only on the shapes of the aggregates but also on the contact areas between the exterior surfaces of the aggregates and the water between the aggregates (Kohne et al., 2009).

Practical modelling is normally interested in large scales, but it is the processes occurring in the pore scale that underpin the phenomena measurable at large scales. Understanding the pore-scale processes is hence important to improve large-scale modelling (Blunt et al., 2013; Porta et al., 2013; Raoof et al., 2013; Tartakovsky et al., 2007; Zhang et al., 2005). Since it is difficult to measure the pore-scale processes due to the opaque nature of the soils, pore-scale modelling in combination with tomography has been increasingly used to bridge this gap. For example, using X-ray computed tomography or focused-ion beam/scanning electron microscopy (FEM/SEM), one can visualise the interior structure of a medium at resolutions as fine as a few nanometres (O'Donnell et al., 2007). These, together with the development in pore-scale models, such as the lattice Boltzmann model, the smoothed particle hydrodynamic model and the pore-network model, have substantially improved our understanding of some fundamental transport processes occurring in porous media (Kang et al., 2010; Raoof et al., 2013; Zaretskiy et al., 2010), which would remain unknown otherwise, such as accumulation of chemical at interface of two materials (Zhang et al., 2010), mixing of reactive solute transport (Scheibe et al., 2013), trapping of chemical by immobile water in structured media (Gouze et al., 2008), and nonlinear increase of the longitudinal dispersivity with saturation (Raoof and Hassanizadeh, 2013).

The purpose of this paper is to use pore-scale modelling and 3D images acquired previously using X-ray computed tomography to directly calculate the mass transfer rate between mobile and immobile waters in structured media under different saturations. The calculated transfer rate was then linked to the memory function commonly used in the literature to simulate chemical transport in structured porous materials. Since the intra-aggregate pores in the materials were smaller than the tomography resolutions, they cannot be explicitly resolved and the solid phase in the images is actually porous aggregates; their impact on solute transport was described by an effective diffusion coefficient. In this work, we assumed that the aggregates were fully saturated, and that water distribution within the inter-aggregate pores was determined by inter-aggregate pore sizes and pore connectedness. In all simulations, the initial concentration in the aggregates was assumed to be zero; the initial concentration in the water between the aggregates was instantly changed from zero to a constant and then remained unchanged, so that we can approximate such a

temporal change in concentration by  $\partial c/\partial t = c_0\delta(t)$ . Solute transport from water between the aggregates into the aggregates was through the water-aggregate interfaces, which in turn depend on saturation as we assumed that there is no water film over the aggregate surfaces due to porous nature of the aggregates. We simulated solute transport under different saturations. The simulated solute concentration and flux at all voxels in the aggregates were spatially averaged, and they were then used to calculate the average mass transfer rate from water between the aggregates into the aggregates. After normalisation, the change of the average transfer rate with time is the memory function. We analysed the memory functions obtained under different saturations and fitted them to an empirical formula. We also discussed and proved how to extend the memory function obtained in such ways to simulate reactive solute transport.

## 2. Background

The transport of non-reactive solute in aggregated soils is often described by the following equation when water within the aggregates can be assumed to be stagnant:

$$\begin{aligned} \frac{\partial \theta_m C_m}{\partial t} &= \nabla D \nabla C_m - \nabla q_m C_m - R, \\ R &= \frac{\partial \theta_{im} C_{im}}{\partial t}, \end{aligned} \quad (1)$$

where  $C_m$  is average concentration of the water between the aggregates,  $\theta_m$  is the mobile water content,  $D$  is hydrodynamic dispersion coefficient,  $q_m$  is water flow rate and  $R$  is the mass transfer rate between the chemical inside and between the aggregates, and  $C_{im}$  and  $\theta_{im}$  are the concentration and water content in the aggregates, respectively. If the aggregates are saturated and the initial concentration in them is zero, the change of the chemical mass inside the aggregates can be described as follows (Cvetkovic, 2012; Gouze et al., 2008; Haggerty and Gorelick, 1995):

$$\frac{\partial (\theta_{im} C_{im})}{\partial t} = \int_0^t \frac{\partial C_m}{\partial \tau} g(t - \tau) d\tau, \quad (2)$$

where  $g(t)$  is called memory function and independent of  $q_m$  and  $D$  for water flow and solute transport through the inter-aggregate pores. Physically, the memory function is the probability that a chemical molecule, which is initially on the aggregate surfaces, enters and stays in the aggregates at time  $t$ ; it depends on geometry and effective diffusion coefficient of the aggregates, and not on water flow and solute transport in the water in the inter-aggregate pores.

The natural aggregates are geometrically complicated, and it is impossible to derive an analytical function to describe the probability  $g(t)$ . Therefore, practical modelling often uses empirical formulae to approximate the memory function. The commonly used formulae include gamma function (Chen and Wagenet, 1995), long-normal function (Russo et al., 2010), and a combination of exponential and power-law functions (Cvetkovic, 2012). Validation of these empirical functions is normally carried out by calibrating them against the observed data from column or field tests.

For a given soil, we can directly calculate its memory function from pore-scale simulating solute transport through the soil structure. Since the memory function is a property of soil structure and independent of water flow and solute transport between the aggregates, for simplification of calculation we assumed that the initial concentration in the aggregates is zero, and the initial concentration in the water between the aggregates instantly increases from zero to  $C_M$  and then remains unchanged. Under these conditions, the temporal change of the concentration in the water between

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