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Large time behaviour of oscillatory nonlinear solute transport in porous media

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HIGHLIGHTS

- Oscillation for nonlinear reacting solute gives nonlinear reaction-diffusion problem.
- Mechanical dispersion coefficient involves the time averaged absolute discharge.
- Nonlinear reactions lead to a large time limit asymmetric, non-Gaussian mixing zone.

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ABSTRACT

Oscillations in flow occur under many different situations in natural porous media, due to tidal, daily or seasonal patterns. In this paper, we investigate how such oscillations in flow affect the transport of an initially sharp solute front, if the solute undergoes nonlinear sorption and, disregarding molecular diffusion, mechanical dispersion. By homogenization, we show that after many cycles, the transport converges to a zero convection, pure nonlinear diffusion problem. The impact of the oscillatory flow requires an adjustment of the dispersion coefficient, where instead of the modulus of the particle velocity, we have to use its time averaged value. Physically, this agrees with a velocity defined by the distance travelled by particles in solution, divided by the time needed for that displacement. The nonlinearity of the sorption reaction is retained in the concentration distribution of the mixing zone, because in the large time limit, this distribution is not Gaussian but asymmetric. With numerical simulations, we show that this convergence may occur relatively fast (say 10 cycles). The implication of the diffusion like large time behaviour is that the transition zone continues to spread beyond the zone of convective oscillation.

1. Introduction

When studying flow and transport in natural porous media in field conditions or in packed columns in the laboratory, steady state flow is often assumed. However, there are many important cases where transient flow needs to be considered (Dagan et al., 1996, Dentz and Carrera, 2003).

A special case of transient conditions is that of oscillating flow, where flow in one direction is compensated by a complete reversal. For conditions studied in soil science and other geosciences, for instance, seasonal fluctuations are often oscillatory. Examples are seasonal wetting and drying, although wetting and drying may occur at different time scales. Atmospheric forcing that has an oscillatory aspect is not limited to precipitation/evapotranspiration cycles, but also related with fluctuating air pressures (Neeper, 2001; Neeper and Stauffer, 2012; Jaeger and Kurzweg, 2003). In fact, oscillatory gas exchange for porous media has been investigated decades ago when Raats and Scotter (1968) considered flow that varies sinusoidally with time and investigated the dispersive behaviour due to such oscillations. The rate of dispersion can be described as a function of the Peclet number and the dimensionless amplitude of displacement, and this was experimentally tested by Scotter and Raats (1968) and elaborated numerically by Scotter and Raats (1969).

More recent is the work on fluctuating interfaces in shallow groundwater by Eeman et al. (2012, 2016) and Cirkel et al. (2015). At a small time scale, daily oscillations may occur at the plant root surface due to the day-night pattern of transpiration (Espeleta et al., 2016). Also at drinking water wells, oscillating

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conditions may be part of management (Pauw et al., 2016) to keep filters open (free from iron oxide deposits) by periodically extracting and discharging water. In underground energy or chemical storage, oscillating conditions may be important, for instance for seasonal underground heat storage and harvesting. In the context of tracer dispersion in estuaries, Kay (1997) investigated oscillating flows due to tidal reversals. Of interest is also the analysis by Pool et al. (2016), who considered oscillatory movement for an inert (conservative) solute. Their mathematical analysis, supported by numerical as well as experimental observations, revealed the impact of oscillations on the width of a mixing zone between two solutions with different concentrations. At large times, a symmetrical, Gaussian, distribution of the concentration front developed and they provided expressions of e.g. the width of the concentration front (which displaces) as a function of time.

Oscillating flow and transport has also been considered in chemical engineering. Though not considering a porous medium, Harvey et al. (2001), Reis et al. (2004) and Zheng and Mackley (2008) investigated mixing in a reactor with oscillatory flow. There is also earlier work for baffled tubes on mixing (Dickens et al., 1989) and heat transfer (Mackley and Stonestreet, 1995) for such flow conditions. Recently, Wang et al. (2017) considered mass transfer for a pulsed disc and doughnut (PDD) extraction column.

As Neeper and Stauffer (2012) observed, the combination of periodic flow of the fluid in the pores, on the long term leads to diffusion type of behaviour, that can be captured with an effective diffusion coefficient. This was also the key point of Cirkel et al. (2015), who combined oscillating flow with cation transport, for the case of nonlinear (Gapon type) cation exchange. The latter speculated, based on an approximation of the governing equations, that the effective diffusion coefficient comprises a generalization of the so-called mechanical dispersion coefficient. Whereas in classical theory the mechanical dispersion coefficient equals the product of a formation factor (the dispersivity) and the absolute value of the pore water velocity, for oscillatory flow the time average of the absolute value of pore water velocity should be used. Therefore, for zero net displacement of a mixing zone, the velocity in the mechanical dispersion term is given by the travelled distance of particles in either direction divided by the time needed for this displacement.

With regard to the work by Cirkel et al. (2015), it is worthwhile to emphasize that nonlinear adsorption/desorption essentially differs from the nonreactive or linear sorption and transport behaviour. As Van Duijn and Knabner (1991, 1992) and Van Der Zee (1991) showed, for appropriate initial and boundary conditions (steady flow), traveling wave behaviour is found, as nonlinear sorption counteracts dispersional spreading. For reversed conditions, sorption and dispersion enhance each other, leading to rapidly spreading fronts: rarefaction waves (RW). For oscillatory flow, Cirkel et al. (2015) showed that these types of waves are not observed after a number of oscillations.

It is the scope of this paper, to reconsider the transport of a nonlinearly reacting (adsorbing/desorbing) solute under an oscillating flow regime and to investigate the large time behaviour of the solute front and mixing behaviour. In particular, we show that at large times, neither TW nor RW spreading is obtained. In fact, the front approaches a pure diffusion/dispersion type of spreading that is non-Gaussian due to the nonlinear reaction, and with an adjusted mechanical dispersion coefficient.

2. Problem statement

We consider a flow field describing an oscillating pore water velocity $V(t)$, with period T and mean $\langle V\rangle = 0$. This flow field transports a reactive solute through an infinitely long and one dimensional column. Solute transport is given by the well-known convection-dispersion equation. In case of nonlinear adsorption of the solute subject to an initial step front, the transport is described by Convection-Dispersion-Reaction Problem (CDRP)

$$\frac{\partial\varphi(u)}{\partial t} + V(t)\frac{\partial u}{\partial x} = D(t)\frac{\partial^2 u}{\partial x^2} \quad x \in \mathbb{R},\ t > 0, \tag{1}$$

$$u(x,0) = \begin{matrix} 1 & x < 0 \\ 0 & x > 0 \end{matrix}; \tag{2}$$

where $u \geq 0$ denotes a scaled solute concentration, the function $\varphi(u)$ is strictly increasing and describes the accumulated solute on a volumetric basis, t is time, x is position, and D is the hydrodynamic dispersion coefficient (Bear, 1972). We assume sorption to be given by the Freundlich expression:

$$\varphi(u) = u + Au^p \quad A > 0, \quad 0 < p < 1. \tag{3}$$

Further, we ignore molecular diffusion, hence

$$D(t) = \alpha|V(t)|, \tag{4}$$

with $\alpha > 0$ denoting the dispersivity. We rewrite (1) as

$$\frac{1}{|V(t)|}\frac{\partial\varphi(u)}{\partial t} + P(t)\frac{\partial u}{\partial x} = \alpha\frac{\partial^2 u}{\partial x^2}, \tag{5}$$

where $P(t)$ is a function given by $V(t)/|V|(t)$, i.e. (Fig. 1),

$$P(t) = \frac{V(t)}{|V(t)|} = \begin{cases} 1 & in\ \{V > 0\}, \\ -1 & in\ \{V < 0\}. \end{cases} \tag{6}$$

Next, we introduce as new time scale

$$\tau = \int_0^t |V(z)|dz, \tag{7}$$

which is the total travelled distance of the fluid particle in time t. With $v(x,\tau) = v(x,\tau(t)) = u(x,t)$ and $P^*(\tau) = P^*(\tau(t)) = P(t)$, we find the transformed problem

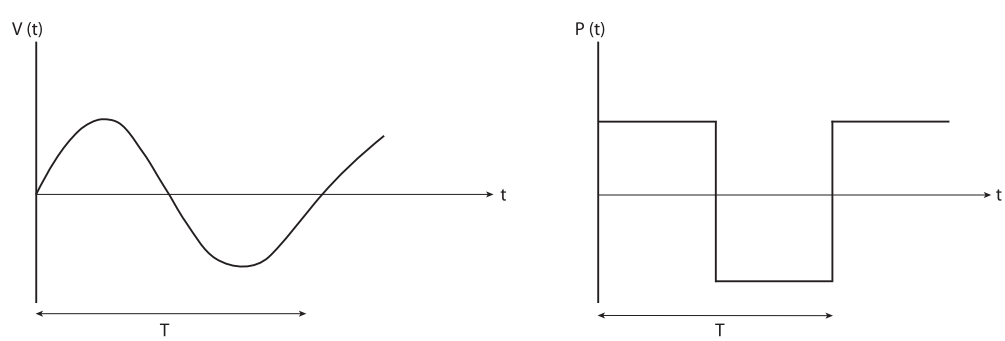

Fig. 1. . Illustration of the velocity as a function of time and the function $P(t)$, where T is the cycle period.