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Evidence of preferential path formation and path memory effect during successive infiltration and drainage cycles in uniform sand columns

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article info abstract

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The formation of preferential flow paths in the partially saturated zone, and in naturally structured media, is well known. This study examines non-uniform flow in uniform sand columns under different pressure and infiltration/drainage conditions. Experiments were carried out in a vacuum box, with applied suction set to three different heads, and with infiltration fixed at two different flow rates. Tailing observed in some conservative tracer breakthrough curves suggests the formation of immobile resident water pockets which slowly exchange mass with the flowing water fraction. The applied suction controlled the degree of water immobilization whereas flow rate had minimal effect on the dynamic behavior. Trapping and exchange of water occurred repeatedly during successive infiltration and drainage cycles, implying a (hysteretic) memory effect of the previously formed preferential flow paths. Flow and solute transport modeling suggests that these dynamics can be described by a mobile–immobile model that corroborates measurements suggesting preferential flow path formation. These findings have implications for the natural attenuation of contaminants in the partially saturated zone, but also for the persistence of a contamination source exposed to repeated conditions of infiltration and drainage.

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1. Introduction

Infiltration events can percolate water from precipitation, irrigation, spills or leakage. Water infiltrating into the subsurface via the partially saturated (or "unsaturated") zone commonly encounters resident "old" water from previous infiltration events. Old water resides in water pockets which are only partially connected to the main flow paths and can interact with the "new" water by exchanging mass and solute. This interaction has strong implications for contaminant transport through the partially saturated zone and it is therefore of particular interest to obtain a better understanding of the process. The variability in

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soil type and structure, vegetation coverage, flow rate, immobile water content and pressure gradients adds to the complexity of the interactions.

Water that moves relatively quickly through the unsaturated zone is termed "preferential flow" and leads to irregular infiltration patterns in the soil profile [\(Hendrickx and Flury,](#page--1-0) [2001](#page--1-0)). In this context, preferential flow paths are conceptualized to conduct highly mobile water, while an immobile fraction is conceptualized to accommodate the more slowly flowing water. This fraction includes relatively isolated regions which are loosely associated with the flow in the vadose zone, such as dead-end pores and intra-aggregate water [\(Delleur, 2006\)](#page--1-0). Among the factors responsible for preferential path formation are the heterogeneity of the porous medium [\(Winiarski et al., 2013\)](#page--1-0), the initial water content ([Kätterer](#page--1-0) [et al., 2001](#page--1-0)), and water repellency properties of the porous medium [\(Ritsema et al., 1993\)](#page--1-0).

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Field studies have shown that contaminants, tracers and nutrients can follow preferential flow paths [\(Gouet-Kaplan](#page--1-0) [et al., 2012\)](#page--1-0). This suggests that solute movement towards the saturated zone can be accelerated or that slow release can take place for a solute that has been trapped for significant time in resident water pockets. Furthermore, the mixing of infiltrating and resident water can induce geochemical reactions between their components which under equilibrium flow conditions would not take place. [Ghodrati and Jury \(1992\)](#page--1-0) investigated the effect of soil structure and irrigation method on pesticide mobility. They found that a significant portion of the pesticides traveled longer distances than expected through loamy sand, despite their high adsorption coefficients. This observation was attributed to flow through preferential flow paths. The same effect was reported by [Gjettermann et al. \(1997\)](#page--1-0), who found that dye tracers follow macropores, mainly earthworm channels; in particular, the authors found that flow through the soil matrix was relatively more significant at lower rates of irrigation. In a field study on grassland soils, [Weiler and Naef \(2003\)](#page--1-0) showed that water bypassed the soil matrix and the less permeable regions under both partially and fully saturated conditions. Such studies also highlight the significance of macropores in runoff generation at a catchment scale. The phenomenon of old water pockets communicating with a network of macropores can explain the hydrological paradox of quick response of storm flow to precipitation with simultaneously introduced conservative tracer concentrations, in which old water is mobilized quickly and effectively dilutes the tracer that marks the precipitation (new) water ([Bishop et al., 2004; Kirchner, 2003](#page--1-0)).

More detailed studies have been carried out at the laboratory scale to examine such phenomena. For example, [Goldreich et al. \(2010\)](#page--1-0) found that infiltration and oven-drying cycles increase the release of the pesticide metolachlor. They attributed this effect to the exchange of water held within the heavy clayey soil aggregates when applying the infiltration– drying cycles. [Haouari et al. \(2006\)](#page--1-0) found that high initial water content in a clayey soil column increases the mobility of the herbicide linuron. This is due to the fact that high initial water content distributed the linuron on the outer region of soil aggregates. In the same study, the application of linuron after infiltration–drying cycles was found to reduce its mobility as the herbicide moved into inter-aggregate regions.

The appearance of preferential paths, even in uniform porous media and at small scales, has been verified by direct observation using sequential Magnetic Resonance Imaging (MRI). Minor differences in local values of hydraulic conductivities were demonstrated to have a significant effect on flow velocities and their direction ([Hoffman et al., 1996\)](#page--1-0). Moreover, MRI measurements provide insight into possible, unexpected small-scale heterogeneities or non-parallel flow fields ([Oswald](#page--1-0) [et al., 1997\)](#page--1-0). Both studies provide unequivocal proof of the existence of preferential flow paths which are normally only inferred from breakthrough tailing and simulations.

While preferential pathways have been the subject of significant research, very few studies have focused specifically on flow and transport dynamics in the context on old and new water, and their interactions. At the field scale, studies investigating old–new water interactions have used conservative chemical tracers and isotope analyses to show that old water can be a significant component in vadose zone outflow into streams or aquifers. This observation has been demonstrated at different time scales varying from several hours [\(Collins et al., 2000;](#page--1-0) [Turton et al., 1995\)](#page--1-0) to days [\(Sklash and Farvolden, 1979](#page--1-0)) or years ([Brooks et al., 2010; Gvirtzman et al., 1988](#page--1-0)) after infiltration of new water.

At the laboratory scale, [Gouet-Kaplan and Berkowitz \(2011\)](#page--1-0) demonstrated through direct visualization that alternating infiltration and drainage cycles of dye tracers through a twodimensional glass lattice micromodel leads to formation of old water pockets. The micromodel experiment served as a proxy for flow through a homogeneous medium and revealed two characteristic regimes. First, old water is drained rapidly by invasion of new water. Second, a near steady-state regime is established in which old water pockets are only partially connected to the main flow paths. A second study ([Gouet-Kaplan et al., 2012\)](#page--1-0) provided evidence of the presence of old water pockets in sand columns exposed to infiltration and free drainage. The two regimes reported in [Gouet-Kaplan](#page--1-0) [and Berkowitz \(2011\)](#page--1-0) were again observed, and it was found that the amount of immobile water depends strongly on the sand particle size and the initial water content. In some cases, more than one third of the old water remained in the sand column after five pore volumes were flushed. A mobile– immobile model (MIM) [\(van Genuchten and Wierenga, 1976](#page--1-0)) for water movement was applied to fit the experimental data (discussed below).

In modeling preferential flow, two-phase models aim to describe the full physics of the interaction between the two phases (i.e., water and air). Infiltration and drainage events induce transient flow conditions in a partially saturated (two-phase) porous medium as the saturation of water and the alternate phase change. Capillary pressure can significantly affect flow distribution in these cases. Several effects are contained within the capillary pressure vs. saturation curve, such as immiscibility, surface tension, fluid viscosity, grain size distribution, and microscale and macroscale heterogeneities ([Hassanizadeh and Gray, 1993](#page--1-0)). In modeling transient flow, pore-network models physically describe processes of phase trapping or interconnectivity which are induced by capillary pressure [\(Joekar-Niasar and Hassanizadeh, 2012\)](#page--1-0). Moreover, the existence of a second phase that becomes trapped is responsible for possible hysteresis effects, which can be taken into account. However, such two-phase models are not widely used to describe larger scale experimental observations.

Instead, for simplicity the one-phase Richards equation is often used in describing flow in partially saturated homogeneous media ([Richards, 1931](#page--1-0)). To model flow through structured media, several approaches have been adopted; most commonly, variations of the dual porosity and dual permeability models have been employed (Šimů[nek and van Genuchten,](#page--1-0) [2008\)](#page--1-0). As far as flow modeling in the vadose zone is concerned, for simplicity, it is common to use single porosity hydraulic models combined with the Richards equation; solute is then treated as being subject to partial retention in the matrix leading to physical non-equilibrium. Such MIM models have been widely used to characterize flow in the unsaturated zone of structured media. However, it is not always possible to distinguish between physical and chemical processes that control the capture and release of a solute. [Nkedi-Kizza et al.](#page--1-0) [\(1984\)](#page--1-0) demonstrated that often-used physical and chemical non-equilibrium transport models (i.e., mobile and immobile model (MIM) and the two-site sorption model, respectively)

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