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Imaging water fluxes in porous media by magnetic resonance imaging using D_2O as a tracer

Andreas Pohlmeier[a,*], Dagmar van Dusschoten[b], Lutz Weihermüller[a], Ulrich Schurr[b], Harry Vereecken[a]

[a]Forschungszentrum Jülich, ICG-4, Jülich 52425, Germany
[b]Forschungszentrum Jülich, ICG-3, Jülich 52425, Germany

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Abstract

In this study, we investigate the usefulness of D_2O as a conservative tracer for monitoring water flux by MRI in a heterogeneous sand column. The column consisted of a cylindrical 3×9-cm packing of fine sand in which an 8-mm diameter cylindrical obstacle was placed. Constant steady-state flux densities between J_w=0.07 and 0.28 cm min^{-1} corresponding to mean pore flow velocities between 0.20 and 0.79 cm min^{-1} were imposed at the top of the sand column, and a constant hydraulic head of −39 cm was maintained at the lower boundary. We injected pulses of 0.01 M $NiCl_2$ and 55% D_2O and monitored the motion of the tracer plumes by MRI using a fast spin echo sequence over a period of 20 min. We observed that the center of gravity of all plumes moved with the mean pore flow velocity, which showed that D_2O behaves as a conservative tracer. The motion of the tracer plume at J_w=0.14 cm min^{-1} was validated by a numerical simulation using HYDRUS2D, which reproduced the experimentally observed behavior very satisfactorily.

Keywords: MRI; Sand; Transport; Flux; Tracer; Modeling; HYDRUS2D

1. Introduction

The understanding of water movement and solute transport in porous media is of huge importance for the prediction of a wide range of transport phenomena in soil science, hydrology and material sciences. In most classical approaches, the water and solute movement is observed in column or field experiments where the breakthrough of the used tracers or substances is studied. Despite the fact that the breakthrough curves are described well by 1D transport models, it has been shown that the spatial distribution of solute in the soil columns cannot be predicted accurately [1]. One reason might be the treatment of the soil columns as black boxes. This drawback may be partly overcome by using invasive probes like time domain reflectometry (TDR) [2] or in situ extraction of the liquid phase [3], which yields information about water content and solute composition at few defined sites inside the column. However, natural porous media may be quite heterogeneous, and therefore a much higher resolution for the monitoring of water and solute transport within the porous body is necessary. As such, noninvasive 2D or 3D imaging methods are essential for a thorough understanding of local water and solute transport. For these purposes, 3D noninvasive imaging methods like X-ray tomography [4,5], ground penetration radar [6], electrical resistivity tomography [7] and magnetic resonance imaging (MRI) [8–12] have been adapted and further developed to soil processes over the last decades. Although MRI offers the possibility to monitor rapid fluxes (order of magnitude: millimeters per second) directly by flow imaging techniques [11], slower flow rates and water fluxes — typical for soils — can only be monitored indirectly by using tracer substances. The most popular tracers are aqueous solutions of paramagnetic ions such as Cu^{2+} or Ni^{2+} [8,12]. These affect the T_1 and T_2 relaxation times and create contrast with respect to regions without tracer. However, these tracers may adsorb at the liquid–solid interface especially at higher pH values of the medium and therefore do not behave conservatively. As an alternative to the commonly used paramagnetic

* Corresponding author. ICG-4, Forschungszentrum Jülich, D-52425 Jülich, Germany.
E-mail address: a.pohlmeier@fz-juelich.de (A. Pohlmeier).

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ions, we have chosen D_2O as a tracer substance. D_2O behaves chemically very similar to H_2O. But its nuclear spin is unity and it is not excited by ^1H rf pulses in an NMR experiment. This makes it a convenient candidate as conservative tracer for transport monitoring by MRI.

Some studies which used D_2O as contrast agent for monitoring water motion in different porous media exist. Locatelli et al. [13] describe a decrease of T_2 [Carr–Purcell–Meiboom–Gill (CPMG)] with an increase of the H_2O/D_2O ratio in 2% $CuCl_2$ solution, whereas the amplitude is proportional to the water content. The decrease is attributed to the dilution of H_2O by D_2O. These mixtures were only used as reference for the measurement of the T_2 relaxation in porous rocks, but not as tracer for liquid motion. An early study employing D_2O as tracer is the investigation of water diffusion into catalyst pellets [14]. Choi et al. [15] used D_2O as tracer for monitoring diffusion into H_2O-saturated hardened cement. They determined T_2 relaxation using CPMG echo trains and extrapolated the signals to time $\rightarrow 0$ by stretched exponential functions. From the temporal evolution of these amplitudes with immersion time, effective diffusion coefficients were finally obtained. A similar study is the determination of the transport of water in bones which is characterized by a diffusion coefficient in the same order of magnitude (10^{-7} cm^2 s^{-1}) as in Refs. [15,16]. D_2O is also a useful tracer for studies in biological samples. Kovar et al. [17] determined tracer uptake rates in cancer tissues of rats. Ilvonen et al. [18] demonstrated different flow velocities in different compartments of wood xylem by using a 3D steady-state sequence (constructive interference in steady state [CISS]) where the contrast is determined by the ratio T_1/T_2. This affects the signal intensity which is proportional to the mole ratio H/D in the solvent. An example for the usage of D_2O as tracer in technical porous media is the study of Feindel et al. [19], who reported rapid D–H exchange in fuel cell electrode membranes which can be used as a tool for monitoring water fluxes in proton exchange membrane fuel cells. The work most related to our study is the determination of spatially resolved flow velocity and dispersion in chromatographic columns by monitoring D_2O tracer pulses by flash and pulsed gradient spin echo NMR. The tracer moved as a curved cloud with Gaussian cross profile in the flow direction, due to the point injection into the column [20].

The above mentioned studies suggest that D_2O might be a convenient MRI tracer for monitoring fluxes in natural porous media. For this purpose, we used a packed sand column containing binary-type heterogeneity and imposed a steady-state water flux as top boundary condition. The binary heterogeneous structure consisted of a cylindrical body embedded in a homogeneous sand matrix. This obstacle forces the water and solute to flow sideways which should be monitorable by MRI. Additionally, the transport of D_2O is compared with a classical paramagnetic tracer ($NiCl_2$). Finally, we will use a numerical model to evaluate the tracer movement through the column on the basis of independently determined hydraulic parameters.

The type of MRI sequence used should fulfill three special conditions for the monitoring of comparably rapid fluxes by tracers: (i) it needs to be fast enough to prevent water displacements during each measurement that exceed the spatial resolution; (ii) the spatial resolution should be sufficiently small to monitor fluxes in heterogeneous structures; and (iii) there should be a monotone relation between the tracer concentration and the NMR signal intensity. For these D_2O experiments, the influence of deuterons on the relaxation processes of normal water should be minimized. The first and second condition can be met by using a so-called fast or turbo spin echo sequence [21]. Here, several echoes are acquired for each excitation in order to quickly map a matrix of voxels. This decreases the scanning time by a factor of 4 to 32. The third condition means that we use a relatively long repetition time between single excitations relative to the T_1 relaxation time, and an echo time (T_E) as short as possible with our hardware. Since addition of D_2O increases both T_1 and T_2, this setup ensures that the systematic error is below 1%. This now means that the amount of local signal loss is directly coupled to the amount of D_2O that has replaced normal water in each voxel.

2. Methods

2.1. Column experiments

For the experiment, a 9.72-cm-long PMMA column with a 3-cm inner diameter was used (Fig. 1). The bottom was

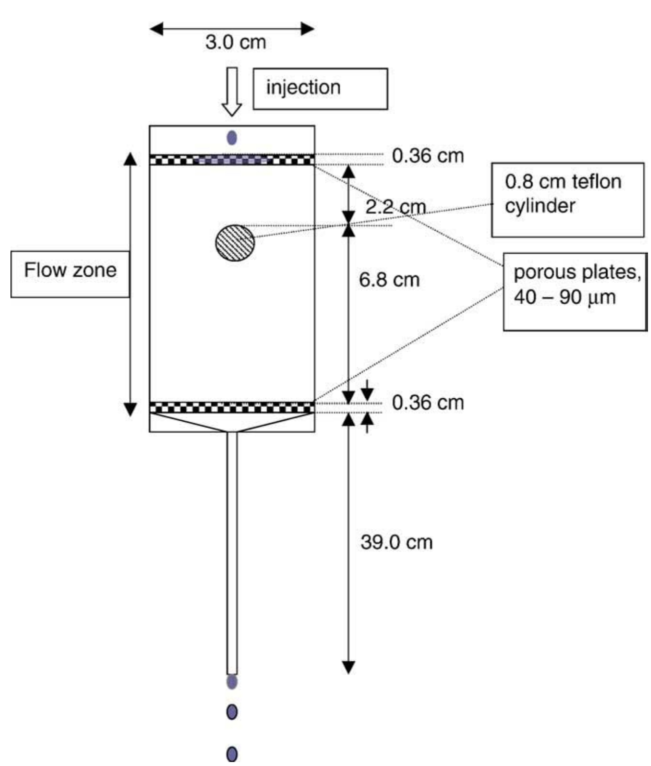

Fig. 1. Sketch of the column used for MRI studies and numerical simulations.