Is breakthrough of solute impacted by the edges of the columns in the case of macropored systems?

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Introduction

In natural systems, preferential flow is the rule rather than the exception. Non-uniform and preferential flows significantly impact mass transport, and by this way most geochemical processes and pollutant dispersion in the environment. Laboratory columns are experimental devices used for monitoring solute transfer through porous media. In particular, several studies used such experimental devices for characterizing mass transfer through heterogeneous systems and macropored systems. However, the design of these devices and its impacts on the experimental results have never been investigated in depth so far, in case of media heterogeneity. In particular, the edge effect is rarely questioned and the transfer is always hypothesized to correspond to a fully developed flow (i.e., flow in an equivalent infinite system). In this study, we question this hypothesis both experimentally and numerically for the case of a macropored system. Tracer elutions, magnetic resonance imaging (MRI), and modeling using multiphysics approaches (FreeFem++) are conducted to demonstrate how flow is affected by edge effects close to the inlet and the outlet of the column, and how the presence of filters (used to prevent particles from exiting the system and clogging the outlet) do impact the flow and solute breakthrough.

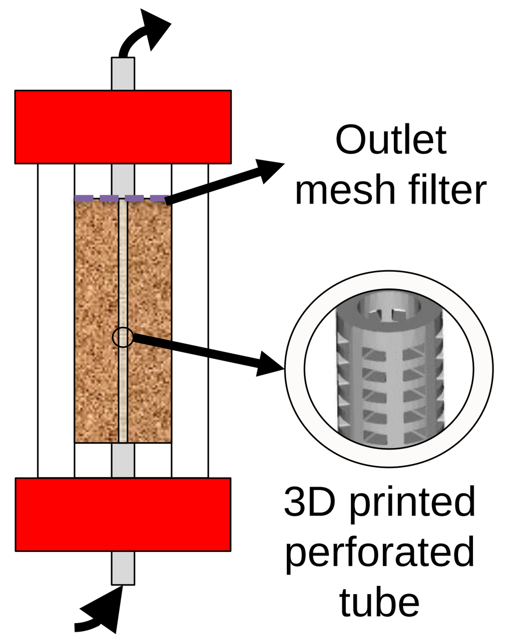
Numerical model

In this numerical model the macroporous and the sandy regions are considered as two porous regions. We modelled both flow equations using Darcy law, and then the solute equation considering the advection-dispersion transport equation.

$$\nabla \cdot (-K\,\nabla p) = 0 \;\; and \;\; \frac{\partial C}{\partial t} + v\nabla C \; = \; \nabla \cdot ([D] \cdot \nabla C)$$

K is the permeability and p the pressure, v the Darcy velocity, C the concentration of the solute and [D] the transversely isotopic dispersion tensor. K takes two different constant values for the macropore and for the porous matrix.

Column experiments

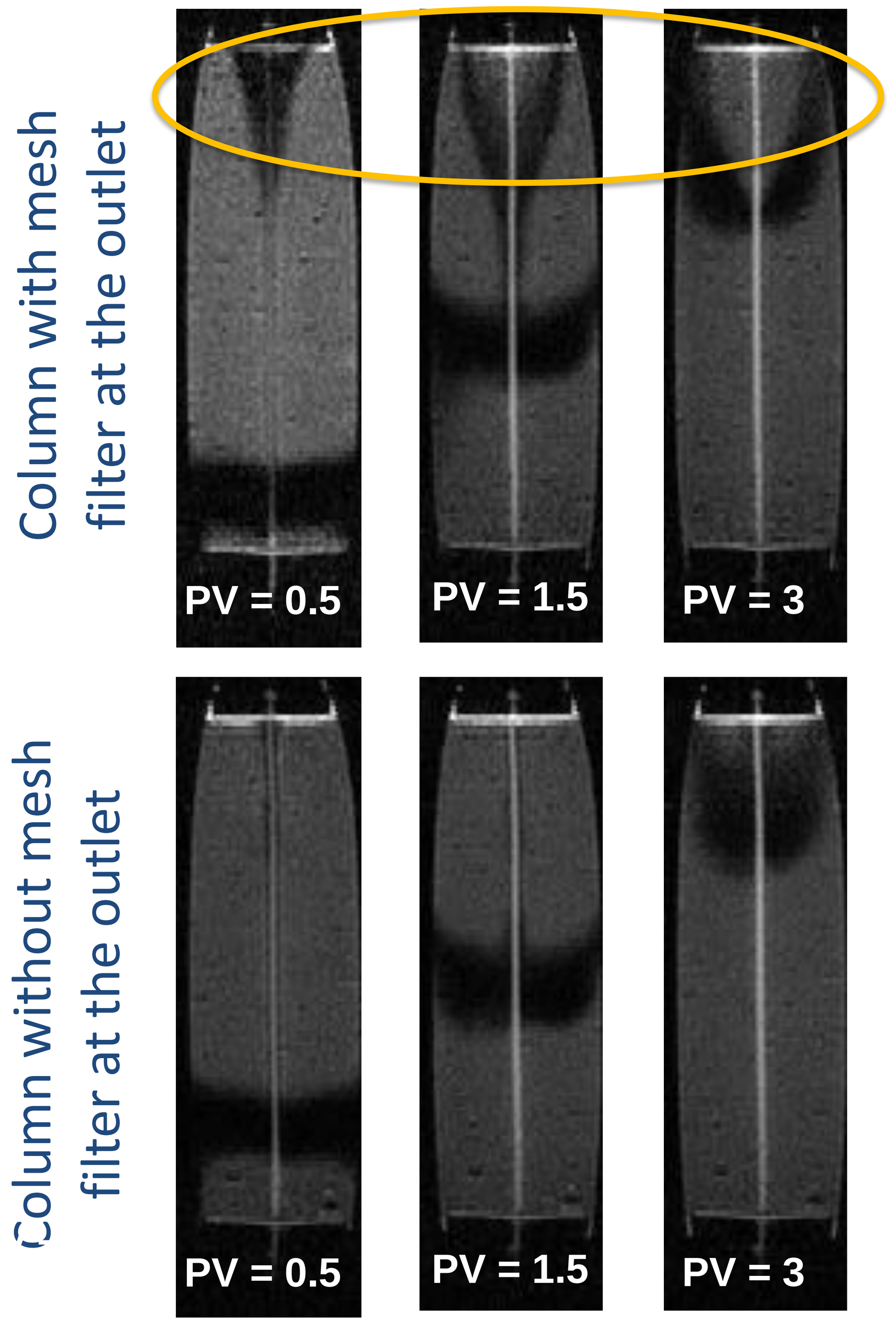


MATERIALS: the experiments were conducted in chromatography column (XK 50 GE Healthcare) with an inner diameter of 5 cm and connected to a chromatography apparatus (ÄKTAprime plus). The column was packed with Hostun quartz sand - D50 ≈ 1 mm - washed with HNO_3 2M. The porous bed height is 15 cm. A perforated tube (i.d. 3 mm) designed with a 3D printer was inserted at the center of the porous bed. The porous volume is around 119 mL. A mesh filter was added or omitted at the outlet to analyze its effect.

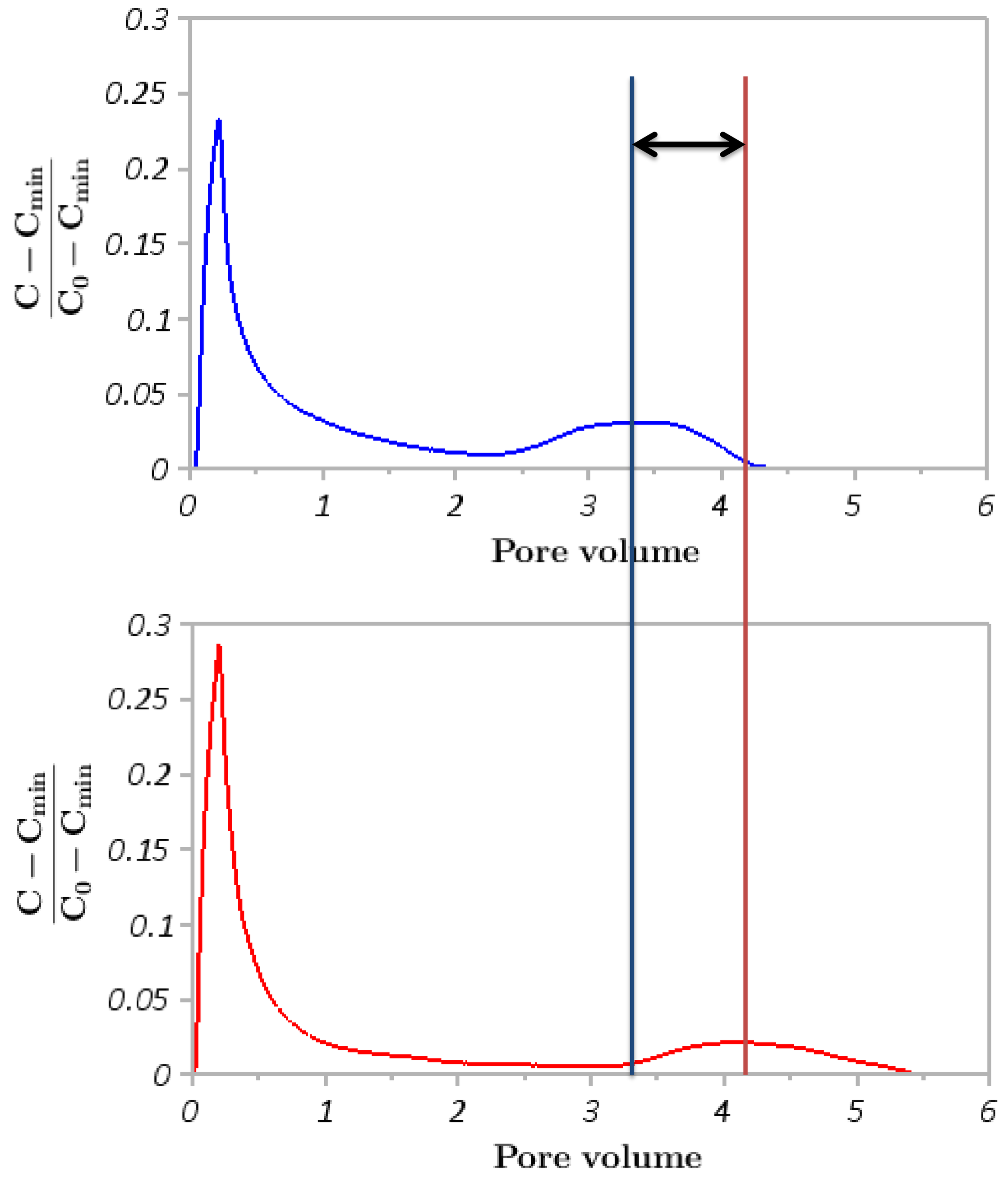
METHODS: $GdCl_3$ 10^{-5} M in ultrapure water is used as eluent and the flowrate is adjusted to Q = 3 mL/min. Injection of 20 mL of $GdCl_3$ (10^{-2} M) is used as ionic tracer of the flow. Concentration of $GdCl_3$ is measured on-line by conductivity. The transfer of $GdCl_3$ within the columns was monitored with a vertical nuclear magnetic resonance spectrometer (DBX 24/80 Bruker), operating with a 0.5 T static magnetic field (20 MHz 1H frequency)

Experimental Results and Modeling

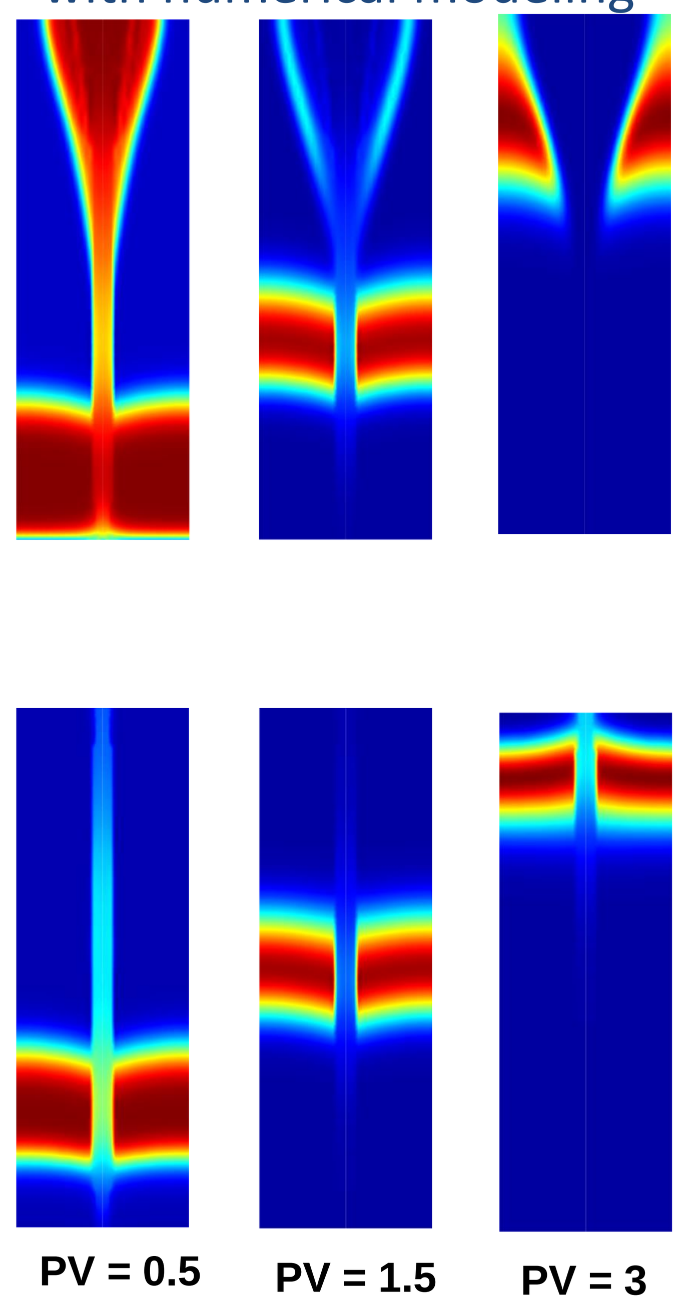
Effect on the flow seen by MRI



Effect on BTCs and residence times in the columns



Identification of the effect with numerical modeling



Conclusions

The shape of the solute BTCs strongly depends on the outlet condition :

- the low permeability of the filter at the column outlet implies a deviation of the flow current lines. This inflection creates an advective exchange from the macropore to the porous medium.
- the absence of a filter also shows a slower elution of the tracer into the sandy matrix.

These observations highlight the influence of outlet conditions on the flows and therefore the need to pay attention to these filters for the design of columns. They also question the relevance of such a system regarding the representativeness of natural porous media.

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