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## Impact of structural heterogeneity on solute transport and mixing in unsaturated porous media: An experimental study

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Solute transport in unsaturated porous media plays a crucial role in soil nutrient dynamics, pesticide leaching, and contaminant migration to aquifers through the vadose zone. Natural porous media are characterized by a strong structural heterogeneity, which impacts solute spreading and mixing and the resulting chemical reaction rates. In addition, incomplete pore-scale solute mixing requires high-resolution experimental measurements to understand the system's mixing dynamics. Our goals are to 1) study the impact of structural heterogeneity on the spatial distribution of fluid phases and 2) establish how fluid phase arrangement impacts solute spreading and mixing during unsaturated flow. We use two-dimensional porous media consisting of circular posts in a Hele-Shaw-type flow cell. The positioning of the posts is random, but we control the medium's heterogeneity by varying the disorder in the posts' diameters and their spatial distribution's correlation length; increasing this length introduces more structure in the porous medium.

In our experiments, we first establish an unsaturated flow pattern with a connected liquid phase and then introduce a fluorescent solute pulse transported by the moving liquid phase. We track the solute concentration and gradients' evolution by taking periodic images of the flow cell and analyzing the fluorescence intensity field. Our results suggest that, as previously shown, decreasing the saturation degree enhances and sustains mixing rates in a disordered porous media due to the emergence of several preferential flow pathways during unsaturated flow. Moreover, increasing the solid posts' spatial correlation reduces the number of air clusters and their interface roughness, and increases their mean size. This leads to fewer preferential flow paths during unsaturated flow for the higher correlated, more structured, porous media, compared to the less structured ones. This reduction in preferential flow paths' number suppresses mixing rate enhancement in the more structured porous media, compared with the less structured porous media, during unsaturated flow. Our experiments show the non-trivial effect of structural heterogeneity and saturation degree on solute mixing in porous media flows. The effects demonstrated by these results are likely to affect reactive solute transport processes such as dissolution and precipitation and adsorption-controlled solute migration.