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Formation of dissolution patterns due to the effect of the coupling of heterogeneity, transient forcing and fluid density variations on mixing in aquifers.

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Solute mixing, spreading and fast dissolution reactions in aquifers are strongly influenced by spatial variability of the hydraulic properties, temporal flow fluctuations and fluid density differences. We study the coupling of heterogeneity, transient forcing and density-driven flow on mixing and chemical reactions between two fluids of different density under a stable stratification. We consider the dissolution of calcite at high Damköhler number, this means under mixing-limited conditions. Two-dimensional constant and variable-density flow and transport Monte Carlo simulations are performed assuming log-normally distributed random permeability fields as well as more complex heterogeneous fields characterized by connected patterns of high and low hydraulic conductivity. While the global mixing and reactivity are on the order of or even smaller than their homogeneous counterparts due to heterogeneity-induced fluid segregation, we find that heterogeneity and transient forcing promote a spatially nonuniform distribution of dispersion and concentration gradients which leads to a strong local enhancement of the mixing and reaction rates. This effect increases with the degree of connectivity of the hydraulic conductivity. We also find that the local maxima of the mixing and reaction rates are localized in regions of strong interface deformation, which correspond to high velocity zones and therefore also large dispersion. When density variations are included there is a decrease in the width of the mixing zone due to the competition between viscous and buoyancy forces. This effect causes a compression of the interface which in turn emphasizes local maxima in mixing and reaction rates. Therefore, the coupling between spatial heterogeneity, density-driven flow and transient forcing magnify geochemical processes and the dissolution efficiency. We also find that the stretching of the interface induced by spatial heterogeneity and transient flow fluctuations coupled with density variations leads to the formation of complex geochemical patterns of reaction hotspots, zones of enhanced reaction efficiency, whose distribution is linked to the medium structure and the deformation properties and topology of the flow field. These results provide new insights into the role of spatial and temporal variability on the mixing and reaction efficiency as well as provide a potential explanation for the formation of complex geochemical patterns observed in karst systems.