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Validity of local equilibrium in heterogeneous formations: Relative importance of mass transfer processes and physical heterogeneity

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Mass transfer processes occur in almost all environments and can seriously affect the behavior of contaminant movement in the subsurface. Complex modeling approaches typically incorporate mass transfer phenomena by describing the system as a dual porosity domain in which particles are exchanged between a mobile and immobile region. Yet, when the characteristic advective time dominates over the characteristic residence time of a solute particle in the immobile domain, the modeling effort can be largely simplified by including an easy-to-use retardation coefficient (local equilibrium models). The validity of local equilibrium models in homogeneous porous media is well known and based on the Damkholer number. Yet, in field applications, natural formations depict a strong variability in the hydraulic parameters that ultimately control the evolution of a solute plume. In this context, we investigate the relative importance of mass transfer processes and physical heterogeneity. To achieve this, we examine the validity of local equilibrium in heterogeneous formations using the small-perturbation approach. Then, we numerically investigate the significance of the latter approximation (mild heterogeneity) by a suite of Monte Carlo transport simulations that consider different models of heterogeneity. Special attention is paid to the role of connectivity.