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Toward a Unified Approach for Quantifying Chemical Transport in Geological Media

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A key problem in subsurface hydrology centers on modelling the physics of chemical transport in heterogeneous geological media, accounting also for reaction mechanisms such as sorption, precipitation and dissolution. Since the early 1950's, dispersive transport in natural porous media has been quantified by the Fickian-based advection-dispersion equation, and stochastic variants thereof, notwithstanding clear and repeated evidence that transport at both field and laboratory scales is non-Fickian. Porous and fractured geological formations have multi-scale heterogeneity, and capturing the complexities of chemical plume migration patterns suggests that, contrary to current practice, small-scale heterogeneities cannot be "averaged out". We have introduced the continuous time random walk (CTRW) framework to provide an effective means to quantify this transport, which allows for transition between non-Fickian and Fickian behavior over a broad range of temporal and spatial scales. The CTRW provides an overarching framework for quantitative modeling, which also encompasses a variety of multirate mass transport, mobile-immobile and fractional derivative equation approaches. Comparison to a wide variety of laboratory- and field-scale observations, and numerical simulations, confirms the relevance and applicability of CTRW theory. Moreover, accounting for chemical reactions between species requires proper treatment of the mixing zone between them, and of fluctuations due to the low concentrations and the localized (pore scale) nature of reactions. Continuum-scale formulations that fully separate effects of transport and reaction result in a significant over-prediction of the quantity of reaction product. We examine these developments and aspects of model application, including parameter estimation, predictive uncertainty, and integration of measurements over a range of scales.