Geophysical Research Abstracts,
Vol. 10, EGU2008-A-02150, 2008
SRef-ID: 1607-7962/gra/EGU2008-A-02150
EGU General Assembly 2008

Continuous time random walk description of non-Fickian transport

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The continuous time random walk (CTRW) framework offers an effective means to quantitatively describe non-Fickian transport in a wide variety of porous and fractured geological formations. The source of non-Fickian behavior is due to the broad spectrum of transition rates that control solute movement. Two principal mechanisms can generate such a spectrum: flow in the complex velocity field of a disordered medium and mass exchange between a mobile phase and a distribution of immobile states. Both of these mechanisms are incorporated into the CTRW framework, which is based on a picture of transport as a sequence of particle transition rates (e.g., between pore spaces, fracture intersections) with displacements s and times t, and the incorporation of the full spectrum of these rates into the transport equations. But even in small-scale, “homogeneous” porous media, subtle and residual pore-scale disorder effects can lead to non-Fickian transport. A central focus of the CTRW approach is an accurate physical model of the entire spectrum $\psi(s,t)$. We find that the uncoupled form, $p(s)\psi(t)$, is an effective representation in many practical situations, although we identify some specific cases where coupled forms of $\psi(s,t)$ are required. We consider solute interaction dynamics as a function of the spectra of advective-diffusive transition times and exchange times, and the relative separation of their respective time domains. We first examine a physical situation in which these two different types of spectra are distinguishable, so that a more complete characterization of the transport can be obtained (i.e., rather than lumping all the rates together). Experimental data are analyzed from a sorbing species transported through a heterogeneous porous domain, and the CTRW is shown to produce excellent fits and predictions, using a single set of parameters. We then focus on observations of non-Fickian transport in two series of sandbox experiments. We represent the main features of the transport in terms of a truncated power-law (TPL), $\psi(t) \sim (t_1 + t)^{-1-\beta} \exp(-t/t_2)$, where t_1 and t_2 are the limits of the power-law spectrum. An excellent fit to the entire BTC data set (including the changes in flow velocity) for each sandbox medium is accomplished with a single set of values of t_1, β, t_2. The use of the full spectrum of $\psi(t)$ is necessary to describe the transition to Fickian behavior, and to account for the dynamics of non-Fickian transport.