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Use of a variable-index fractional-derivative model to capture transient dispersion in heterogeneous media

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ABSTRACT

Field and numerical experiments of solute transport through heterogeneous porous and fractured media show that the growth of contaminant plumes may not exhibit constant scaling, and may instead transition between diffusive states (i.e., superdiffusion, subdiffusion, and Fickian diffusion) at various transport scales. These transitions are likely attributed to physical properties of the medium, such as spatial variations in medium heterogeneity. We refer to this transitory dispersive behavior as "*transient dispersion*", and propose a variable-index fractional-derivative model (FDM) to describe the underlying transport dynamics. The new model generalizes the standard constant-index FDM which is limited to stationary heterogeneous media. Numerical methods including an implicit Eulerian method (for spatiotemporal transient dispersion) and a Lagrangian solver (for multiscaling dispersion) are utilized to produce variable-index FDM solutions. The variable-index FDM is then applied to describe transient dispersion observed at two field tracer tests and a set of numerical experiments. Results show that 1) uranine transport at the small-scale Grimsel test site transitions from strong subdispersion to Fickian dispersion, 2) transport of tritium at the regional-scale Macrodispersion Experimental (MADE) site transitions from near-Fickian dispersion to strong superdispersion, and 3) the conservative particle transport through regional-scale discrete fracture network transitions from superdispersion to Fickian dispersion. The variable-index model can efficiently quantify these transitions, with the scale index varying linearly in time or space.

1. Introduction

Anomalous or non-Fickian transport has been well documented by the hydrology community, motivating the development of various nonlocal transport theories, such as the Stochastic Averaging of the classical Advection–Dispersion Equation (SA-ADE) method (Cushman, 1987; Neuman and Tartakovsky, 2009), the Multi-Rate Mass Transfer (MRMT) model (Haggerty and Gorelick, 1995), the Continuous Time Random Walk (CTRW) framework (Berkowitz et al., 2006), and the Fractional-Derivative Model (FDM) (Baeumer and Meerschaert, 2010; Benson, 1998; Huang et al., 2006; Zhang et al., 2009a). All these approaches imply that the subsurface transport for a solute may not stay at one single diffusive state (i.e., following the same scaling rule) all the time, but rather could transfer between states consisting of superdiffusion, subdiffusion and normal diffusion. The latter behavior is called "*transient dispersion*" in this study. Transient dispersion is a generalization of the transient anomalous dispersion proposed by Meerschaert et al. (2008), who found and quantified the transition from subdiffusion to normal diffusion.

Different transport theories, however, are formulated for specific transition sequences and are therefore valid for different types of transient dispersion. For example, the SA-

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ADE method assumes Fickian dispersion at the Darcy scale before the transport approaches non-Fickian at the regional scale, due to the evolution of medium heterogeneity that a tracer particle experiences during its travel. Contrarily, the CTRW framework describes non-Fickian dispersion at the Darcy scale, before the transport approaches its asymptote (such as the asymptotic Gaussian state) (de Anna et al., 2013). A recent extension of the FDM by Meerschaert et al. (2008) characterizes transient anomalous dispersion from non-Fickian to Fickian observed in many geophysical processes. All the above transport theories have been applied successfully to describe transport processes, as reviewed by Berkowitz et al. (2006) and Neuman and Tartakovsky (2009), respectively. Note that we are not debating that one theory is superior to the other. Rather, we believe that all the above transport theories are applicable for certain conditions, as indicated by their successful applications. Hence the challenging question is: how to develop a physical approach that is general enough to capture all possible transient dispersion sequences predicted by the above models? This question serves as the primary motivation for this study.

Several possible mechanisms may be responsible for the occurrence of, and the transition between, diffusion states. First, normal diffusion typically occurring at specific spatio-temporal scales can transfer to, or be transferred from, any other non-Fickian diffusion depending on the properties of the medium and tracer. For example, Brownian motion is assumed for tracer particles moving in a Darcy-scale homogeneous medium (Neuman and Tartakovsky, 2009), or at a long travel distance much larger than the maximum correlation scale of heterogeneity in a stationary medium (due to the central limit theory). For a nonstationary medium, however, the asymptotic Fickian diffusion may never be reached since the characteristics of particle jumps and/or waiting times evolve with the travel scale (Meerschaert and Scheffler, 2001). In this case, the ending state is anomalous diffusion.

Second, superdiffusion may be space dependent, where the scaling rate may rely on local aquifer properties. The most common condition for fast transport is a preferential flow path that the solute particle can access. The scaling rate or the displacement strength can be either enhanced (when the flow becomes more channelized along the solute particle trajectory) or decreased (when low-permeable deposits surround and separate the flow), depending on the plume position in the host rock and architecture of the medium (Zhang et al., 2007). To account for the local variation of tracer scaling rates, the transport parameters should be conditioned on local aquifer properties, or alternatively, the diffusion state can be adjusted directly.

Third, subdiffusion due to mass exchange between high- and low-permeable depositional zones may be both time and space dependent. For example, laboratory experiments (Dean and Reimus, 2008) showed that mass exchange exhibits different trends with the growth of observation time. Note that the FDM considered in this study assumes independent and identically distributed jumps and waiting times which are different from the correlated CTRW model proposed by Le Borgne et al. (2011) (where their numerical Lagrangian velocity exhibits long-term correlation), although all these models can describe the similar transition from non-Fickian to Fickian transport due to the trapping of solute particles in low-velocity zones. Subdiffusion can converge to normal diffusion at a short travel distance, although the convergence rate can be slow (Berkowitz et al., 2006). This transition occurs because tracer particle residence times (in immobile blocks with a finite size) typically have an upper limit. When transport times exceed this limit, subdiffusion converges slowly to its Gaussian asymptote. A similar transient dispersion can also be found for transport through alluvial aquifer/aquitard systems (Zhang and Meerschaert, 2011). Solute particles released initially in high-permeability ancient channel deposits spread first as normal diffusion, due to the single and relatively homogeneous channel hydrofacies. At a time scale $t \approx l^2/D^*$ (where l is the channel thickness and D^* is the molecular diffusion coefficient), an increase of D^* causes Taylor dispersion effect (Taylor, 1954), where more particles can diffuse vertically into the surrounding low-permeability floodplain deposits and enhance subdiffusion. When the time is much larger than the diffusive time scale $t > l^2/D^*$ (where L denotes the maximum thickness of floodplain layers), anomalous transport approaches Fickian. See also Kitanidis (1988) and Attinger et al. (1999) for the similar diffusive time scale for transport in heterogeneous media to reach the asymptotic Fickian.

It is also noteworthy that superdiffusion due to fast motions and subdiffusion due to mass exchange can occur simultaneously, if preferential flow paths and stagnant flow zones co-exist. Hence the physical model should be able to characterize not only superdiffusion or subdiffusion, but also their mixture. In addition, a transient flow condition can force the anomalous dispersion to vary in time. Both the superdiffusion and subdiffusion therefore can be sensitive to time.

We develop a stochastic model that is general enough to account for all the above possible mechanisms of transient dispersion, and test the above hypotheses using field observations and numerical experimental data. The structure of the remaining paper is organized as follows. In Section 2, a variable-index FDM is proposed to capture all types of transient dispersion discussed above. Numerical solutions are then developed. In Section 3, we investigate the applicability of variable-index FDM in describing the experimental data at several sites. In Section 4, we discuss the possible relationship between model parameters and medium properties. Conclusions are presented in Section 5.

2. Variable-index fractional-derivative model

2.1. Methodology development

The variable-index fractional derivative, which is also called the variable-order fractional derivative (Chechkin et al., 2005; Coimbra, 2003; Samko and Ross, 1993), depicts the influence of time-dependent memory and variable spatial correlation of medium heterogeneity on tracer dynamics (Sun et al., 2009, 2011). The variable-order time fractional derivative of the Caputo type can be written as

$$\frac{\partial^{\alpha(t)} f(t)}{\partial t^{\alpha(t)}} = \frac{1}{\Gamma(1-\alpha(t))} \int_0^t (t-\tau)^{-\alpha(t)} f'(\tau) d\tau, 0<\alpha(t)<1. \quad (1)$$

If the term $(t-\tau)^{-\alpha(t)}$ on the right-hand side of Eq. (1) is replaced by $(t-\tau)^{-\alpha(t-\tau)}$, the updated definition describes