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Tempered anomalous diffusion in heterogeneous systems

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[1] Passive tracers in heterogeneous media experience preasymptotic transport with scale-dependent anomalous diffusion, before eventually converging to the asymptotic diffusion limit. We propose a novel tempered model to capture the slow convergence of sub-diffusion to a diffusion limit for passive tracers in heterogeneous media. Previous research used power-law waiting times to capture the time-nonlocal transport process. Here those waiting times are exponentially tempered, to capture the natural cutoff of retention times. The model is validated against particle concentrations from detailed numerical simulations and field measurements, at various scales and geological environments. **Citation:** Meerschaert, M. M., Y. Zhang, and B. Baeumer (2008), Tempered anomalous diffusion in heterogeneous systems, *Geophys. Res. Lett.*, *35*, L17403, doi:10.1029/2008GL034899.

1. Introduction

[2] Upper truncated power-law distributions have been observed for many geophysical processes at various scales, including interplanetary solar-wind velocity and magnetic-field fluctuations measured in the inner heliosphere [*Bruno et al.*, 2004] and long-term retention of contaminants in alluvial aquifers [*Zhang et al.*, 2007]. To understand the implication of truncation on the dynamics of particle movement, a novel kinetic model is needed [*Metzler et al.*, 2008]. This understanding is critical in practical applications, such as the design of remediation strategies, or the assessment of groundwater quality sustainability. Although this study focuses on long-term transport of passive tracers, we expect that our results may also be useful for other fractal processes, where spatial/temporal upper truncations result from the practical application of power law models to real problems in geophysics. Auxiliary materials[1].

[3] The tempered anomalous diffusion (TAD) model presented here contains both the classical advection-dispersion model and the time-fractional advection-dispersion model [*Schumer et al.*, 2003] as end members. As time passes, the particle plume transitions smoothly from the (pre-asymptotic) fractional to (asymptotic) classical shape, at a rate governed by the truncation parameter. The pre-asymptotic transport is sub-diffusive. A multi-rate mass transport formulation is employed to distinguish mobile and immobile phases.

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2. Methodology

[4] Classical advection dispersion models govern the transition densities of Brownian motion with drift. The space-fractional advection dispersion model for anomalous superdiffusion governs the transitions of a stable Lévy motion with drift. The superdiffusive spreading is the result of large power-law jumps for a moving particle, and the order of the fractional derivative in space coincides with the power-law tail index of the jump distribution [*Meerschaert et al.*, 1999]. Simulations of the underlying process form the basis for particle tracking solutions [*Zhang et al.*, 2006]. Time-fractional advection dispersion models employ stable waiting times, and the long power-law waiting time distribution creates a subdiffusive effect [*Meerschaert et al.*, 2002]. The order of the fractional time derivative codes the power-law waiting time distribution.

[5] Truncated stable Lévy flights were proposed by *Mantegna and Stanley* [1994] to censor arbitrarily large jumps, and capture the natural cutoff present in real physical systems. Exponentially tempered stable processes were proposed by *Cartea and del-Castillo-Negrete* [2007] and *Rosiński* [2007] as a smoother alternative, without a sharp cutoff. In this paper, we temper the waiting times between particle jumps, since real world considerations argue against the possibility of arbitrarily long waiting times with a pure power-law distribution. Sub-diffusive transport has been widely documented in the hydrologic literature [e.g., *Haggerty et al.*, 2000]. Hence the model developed here is expected to have broad applicability.

[6] Multi-rate mass transport typically refers to a system of deterministic equations that relate 1) concentrations in mobile and immobile phases to the divergence of flux in the mobile phase and 2) the transfer of solute mass between phases [*Haggerty et al.*, 2000]. If the solute pulse with total mass m_0 is initially placed in the mobile phase, then the transport equations for the mobile concentration C_M and total (mobile plus immobile) concentration C are [*Schumer et al.*, 2003]:

$$\frac{\partial C_M}{\partial t} + \beta \frac{\partial C_M}{\partial t} \star g(t) = L_x C_M - \beta g(t) m_0 \delta(x) \qquad \text{(1a)}$$

$$\frac{\partial C}{\partial t} + \beta \frac{\partial C}{\partial t} \star g(t) = L_x C; \qquad C(x,0) = m_0 \delta(x) \qquad \text{(1b)}$$

where $\star\ g(t)$ denotes convolution with some memory function, β governs the mobile fraction, $\delta(\cdot)$ is the Dirac delta function, and $L_x = -v\frac{\partial}{\partial x} + B\frac{\partial^2}{\partial x^2}$ drives the advection dispersion process (where v is the velocity and B is the dispersion coefficient). Focusing on the mass-preserving

[1]Auxiliary materials are available at ftp://ftp.agu.org/apend/gl/2008gl034899.

equation for total concentration, we note that a power law memory function $g(t) = \frac{1}{\Gamma(1-\gamma)} t^{-\gamma}$ leads to the time-fractional diffusion equation, since the fractional derivative is a convolution with a power law [*Schumer et al.*, 2003]. Taking Fourier-Laplace transforms in (1b) and solving for $C(k, s) = \int_0^\infty e^{-st} \int e^{-ikx} C(x, t)\, dx\, dt$ yields

$$C(k,s) = \frac{m_0(1 + \beta g(s))}{s + \beta s g(s) + v(ik) + Bk^2}. \qquad (2)$$

The time-fractional case is $g(s) = s^{\gamma-1}$. Replacing $g(t)$ by $g(t/t_0)$ renders both β and g dimensionless.

[7] Tempered anomalous subdiffusion assumes a specific memory function for exponentially tempered power-law waiting times in the immobile phase: $sg(s) = (\lambda + s)^\gamma - \lambda^\gamma$, where the tempering parameter $\lambda > 0$ has units $[T^{-1}]$. Note that the end-member $\lambda = 0$ recovers time-fractional subdiffusion. Invert to discover the memory function:

$$g(t) = \int_t^\infty e^{-\lambda r} \frac{\gamma\, r^{-\gamma-1}}{\Gamma(1-\gamma)} dr \qquad (3)$$

with units $[T^{-\gamma}]$. Rearrange the transform equation (2) to get $sC(k, s) - m_0 + \beta s g(s) C(k, s) = (-v(ik) + B(ik)^2) C(k, s) + m_0 \beta g(s)$. Then invert, using the fact that $f(s - \lambda)$ is the Laplace transform of $e^{\lambda t} f(t)$, to reveal the total concentration equation for TAD:

$$\frac{\partial C}{\partial t} + \beta e^{-\lambda t} \frac{\partial^\gamma}{\partial t^\gamma}\left(e^{\lambda t} C\right) - \beta \lambda^\gamma C = L_x C + m_0 \beta g(t) \delta(x), \qquad (4)$$

where $\frac{\partial^\gamma}{\partial t^\gamma}$ is the Riemann-Liouville fractional derivative, equivalent to multiplying the Laplace transform by s^γ, and β has units $[T^{\gamma-1}]$. One can also write (4) with $\beta_0 = \beta\gamma\lambda^{\gamma-1}$ the dimensionless capacity coefficient, representing the long-term ratio of immobile versus mobile particles.

[8] Rewrite (2) in the form

$$C(k,s) = m_0 \int_0^\infty e^{-uL(k)} \cdot (1 + \beta g(s)) e^{-u(s+\beta s g(s))} du \qquad (5)$$

where $L(k) = (v(ik) + Bk^2)$, and invert to obtain

$$C(x,t) = m_0 \int_0^\infty p(x,u) \cdot q(u,t) du \qquad (6)$$

where

$$p(x,u) = \frac{1}{\sqrt{4\pi B u}}\, e^{-\frac{(x-vu)^2}{4Bu}} \qquad (7)$$

is readily identified as the density of a Brownian motion with drift, $A(u)$. To understand the remaining term $q(u, t)$ in (6), let $D(u)$ be a stochastic process whose density has Laplace transform $f(s, u) = e^{-u(s+\beta s g(s))}$, and define the inverse process $E(t)$ as the smallest $u > 0$ for which $D(u) > t$. Then the density $q(u, t)$ of $E(t)$ is the u-derivative of $P(E(t) \leq u) = P(D(u) \geq t)$. Taking Laplace transforms shows that

$$q(u,s) = -\frac{d}{du}\frac{1}{s} e^{-u(s+\beta s g(s))} = (1 + \beta g(s)) e^{-u(s+\beta s g(s))}$$

in agreement with (5), revealing that the total concentration $C(x, t)$ in (6) is also the probability density of the process $A(E(t))$. Hence, a particle tracking code can be used to solve the equation (4).

[9] Particle tracking is superior to classical Eulerian solvers for large flow systems with complex geometries, and is crucial for inhomogeneous systems that lack exact analytical solutions [*Zhang et al.*, 2006]. An efficient particle tracking solution for equation (4) tracks $x = A(u)$ at operational time $u = E(t)$ via the equivalent relation $t = D(u)$, since the Markov process $D(u)$ is easier to simulate than its non-Markovian inverse [*Meerschaert and Scheffler*, 2008]. Let $D_\lambda(u)$ denote a standard tempered stable process whose density has Laplace transform $e^{-u[(\lambda+s)^\gamma - \lambda^\gamma]}$ as in the work by *Cartea and del-Castillo-Negrete* [2007]. Then it is easy to check that $D(u) = u + D_\lambda(\beta u)$ has a density with Laplace transform $f(s, u) = e^{-u(s+\beta s g(s))}$. Simulation of a tempered stable random variable $D_\lambda(u)$ can be accomplished as follows: Simulate $X(u)$ stable with Laplace transform e^{-us^γ}, using the method of *Chambers et al.* [1976]. Draw an exponential random variable Y with mean λ^{-1}. Use $X(u)$ if $X(u) < Y$, and otherwise, draw both random variables again until the condition $X(u) < Y$ is satisfied. This exponential rejection method is exact (B. Baeumer and M. M. Meerschaert, Tempered stable Lévy motion and transient super-diffusion, submitted to *Journal of Computational Physics*, 2008). Subdivide operational time into small increments of length du, and simulate $x_i = A(u_i) = \sum dA(u)$ and $t_i = D(u_i) = \sum dD(u)$ using the Langevin approach of *Zhang et al.* [2006]. The particle trace $\{(t_i, x_i): i = 0, 1, 2, 3, \ldots\}$ follows the path of the process $x = A(E(t))$, so a histogram of these particle locations gives a numerical solution to equation (4).

[10] In many practical applications, only the mobile mass concentration $C_M(x, t)$ is observable. Take Fourier-Laplace transforms in the mobile equation (1a) to see that $C_M(k, s) = m_0/(s + \beta s g(s) + v(ik) + Bk^2)$. Rearrange to get $sC_M(k, s) - m_0 + \beta s g(s) C_M(k, s) = (-v(ik) + B(ik)^2) C_M(k, s)$, and invert to reveal the mobile equation for TAD:

$$\frac{\partial C_M}{\partial t} + \beta e^{-\lambda t} \frac{\partial^\gamma}{\partial t^\gamma}\left(e^{\lambda t} C_M\right) - \beta \lambda^\gamma C_M = L_x C_M \qquad (8)$$

where $C_M(x, 0) = m_0 \delta(x)$. We can also write $C_M(k, s) = m_0 \int_0^\infty e^{-uL(k)} \cdot e^{-u(s+\beta s g(s))} du$ which inverts to

$$C_M(x,t) = m_0 \int_0^\infty p(x,u) \cdot f(t,u) du. \qquad (9)$$

[11] Invert the Laplace transform $f(s, u) = e^{-u(s+\beta s g(s))}$ to identify the density

$$f(t,u) = e^{-\lambda(t-u) + u\beta\lambda^\gamma} g_\gamma\left((t-u)(u\beta)^{-1/\gamma}\right)(u\beta)^{-1/\gamma} \qquad (10)$$

of a tempered stable process $D(u) = u + D_\lambda(\beta u)$. Here $g_\gamma(t)$ is a stable density with index $0 < \gamma < 1$ and Laplace transform $g_\gamma(s) = e^{-s^\gamma}$, which can be efficiently computed [*Nolan*, 1997]. Now substitute (7) and (10) into (9) for an exact analytical solution of the governing equation (8) for

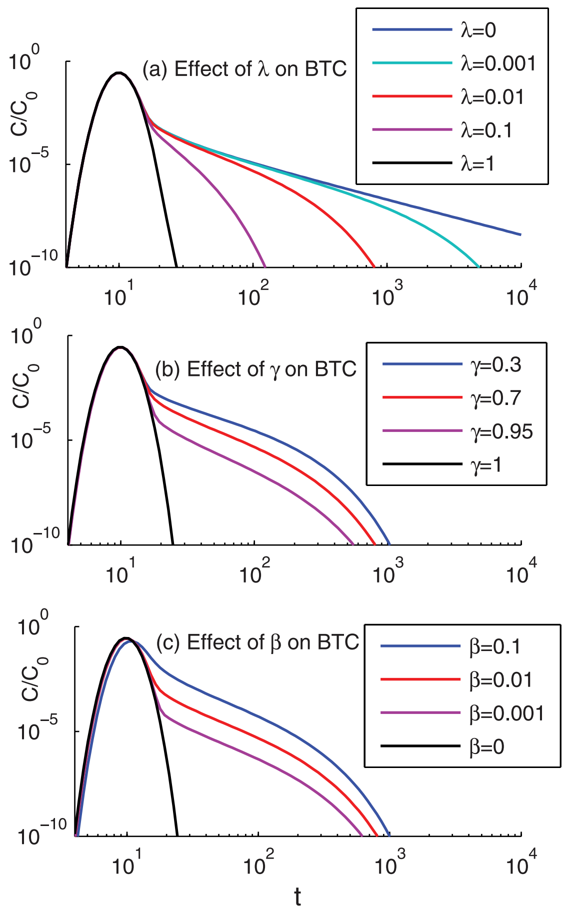

Figure 1. Mobile concentration or first passage time (FPT) at x = 10 of TAD with v = 1 and D = 0.1: (a) we vary λ, holding γ = 0.7 and β = 0.05; (b) we hold λ = 0.001 and β = 0.05; and (c) we keep λ = 0.001 and γ = 0.7.

the TAD mobile concentration. Figure 1 1illustrates the effect of varying the TAD model parameters γ, λ, and β.

[12] To develop a particle tracking solution to the mobile equation (8), first consider a thought experiment, where we solve (8) in the case of pure advection $L_x = -v\frac{\partial}{\partial x}$ at unit speed v = 1. Now $x = A(u) = u$, so that the total particle concentration $C(u, t)$ is equivalent to the probability density $q(u, t)$ of $u = E(t)$. Since the mobile velocity v = 1, the density of mobile particles (flux) crossing the boundary $x = u$ is $C_M(u, t) = \frac{d}{dt}\int_u^\infty C(y, t)\,dy = \frac{d}{dt}P(E(t) \geq u) = \frac{d}{dt}P(D(u) \leq t) = f(t, u)$, the density of $t = D(u)$. Since the same function $f(t, u)$ appears in the general solution (9), we can use this approach to segregate mobile and immobile particles for any advection dispersion operator $L_x = -v\frac{\partial}{\partial x} + B\frac{\partial^2}{\partial x^2}$. Recall that the discretized particle trace is (t_i, x_i) where $x_i = A(u_i)$ and $t_i = D(u_i)$, and that $D(u) = u + D_\lambda(\beta u)$. Write $dt_i = du_i + dD_\lambda(\beta u_i)$ where the first term represents the mobile portion, and the second term (pure jump term) gives the immobile portion. Count the particle as mobile during the time interval du_i and as immobile during the waiting time $dD_\lambda(\beta u_i)$. Then $t = D(u)$ maps mobile time u to clock time t by adding the accumulated waiting time in the immobile zone, and $u = E(t)$ converts clock time to mobile time, which explains why $C(x, t)$ is the probability density of $x = A(E(t))$: The inner process $u = E(t)$ measures the mobile time during which the particle experiences advection and dispersion according to the outer process $x = A(u)$. Differentiating the Laplace transform of the density shows that $D_\lambda(\beta u)$ has mean $\beta u\gamma\lambda^{\gamma-1}$, and this explains why the dimensionless capacity coefficient $\beta_0 = \beta\gamma\lambda^{\gamma-1}$ gives the long-term ratio of immobile versus mobile particles.

[13] The model of *Saichev and Utkin* [2004] with exponentially tempered power-law waiting times is asymptotically equivalent to TAD as $t \to 0$ or $t \to \infty$. The retarding subdiffusion model of *Chechkin et al.* [2002] transitions from a higher fractional order to a lower order. The accelerating subdiffusion model of *Sokolov et al.* [2004], *Sokolov and Klafter* [2005] produces the opposite effect, similar to TAD but remaining mildly subdiffusive at late time.

3. Applications

[14] The numerical simulations of *Zhang and Lv* [2007] demonstrate that the movement of passive tracers through a uniform, two-dimensional, pore-scale, porous medium (where all pores are hydraulically connected) is anomalous (sub-diffusive), as shown by the circles in Figure 2a. The relatively heavy trailing edge of plume snapshots for total concentration [M/L^3] is underestimated by the classical ADE model, but it can be captured by the TAD model (4) for total concentration with a relatively large cutoff parameter $\lambda = 1.0$ [$v = 13.8$, $B = 8.6$, $\gamma = 0.82$, $\beta = 0.035$, $m_0 = 6.6$]. The large λ may be due to the simple uniform medium. *Zhang and Lv* [2007] comment that natural heterogeneous media may exhibit a much longer persistence of anomalous diffusion, implying a smaller λ.

[15] Dye tracer concentration measured along different reaches of the Missouri River between Sioux City, Iowa, and Plattsmouth, Nebraska (regional-scale, ~200-km-long) exhibits a slow decay rate at late time (see symbols in Figure 2b) that is overestimated by the single-rate mass transfer model [*Deng et al.*, 2004]. Since mass loss is negligible, a total concentration [ppb] model is appropriate. As explained by *Deng et al.* [2004], a single transfer rate or a similar storage zone model may not sufficiently capture the complex mass transfer processes, attributed to the wide spectrum of dead-zones in natural rivers and streams (such as reverse flows induced by bends and pools, side pockets, zones between dikes, turbulent eddies, etc). The TAD model (4) with a relatively small $\lambda = 0.01$ hr^{-1} captures the slow decay of observed concentration [v = 5.69 km/hr, B = 2.3 km^2/hr, γ = 0.90, β_0 = 0.03, m_0 = 72.0 micrograms]. Since the observation time is too short to capture the complete behavior, the fitting of λ has high uncertainty. Recall that the dimensionless capacity coefficient $\beta_0 = \beta\gamma\lambda^{\gamma-1}$.

[16] The regional-scale (900 m-long) Lawrence Livermore National Laboratory (LLNL) aquifer system is dominated by fine-grained sediments, where the volumetric proportion of low permeability floodplain facies is as high as 56%. The Monte Carlo numerical BTC for dimensionless concentration C/C_0 produced by *Zhang et al.* [2007] has a heavy late tail, transitioning from power law to exponential (see symbols in Figure 2c), which can be fitted by the mobile TAD model (8) with $\lambda = 0.0006$ yr^{-1} [v = 17.5 m/yr, B = 90 m^2/yr, γ = 0.61, β_0 = 11.9, m_0 = 1]. The original time-fractional advection dispersion model without cutoff

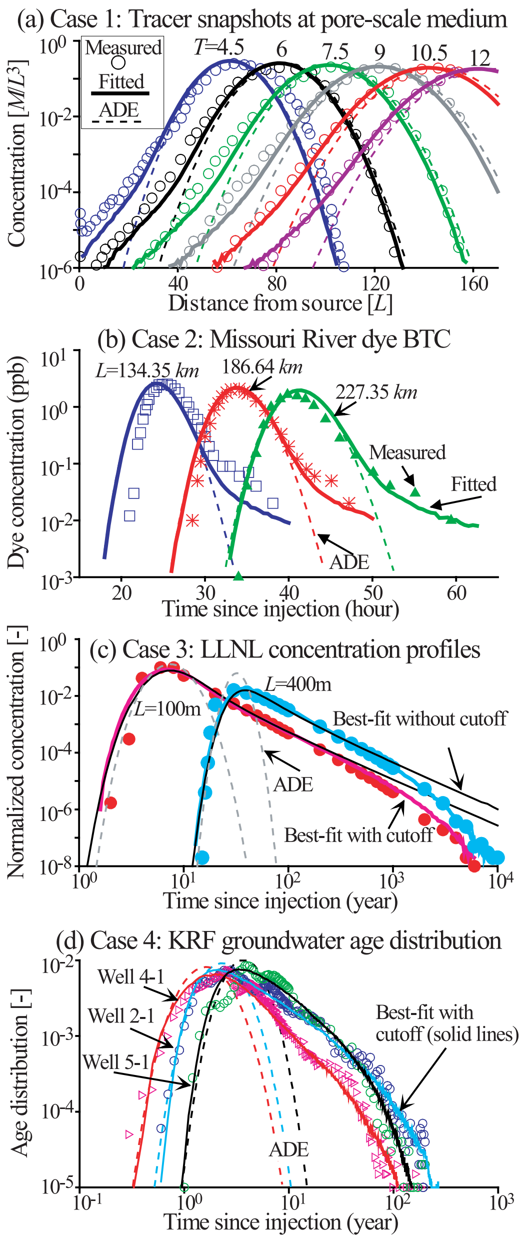

Figure 2. The best-fit concentrations using the TAD model (lines) versus the original plumes from the literature (symbols). (a) The best-fit total concentration snapshots versus the snapshots generated by *Zhang and Lv* [2007] for passive tracer transport through a pore-scale medium. T denotes the transport time. (b) The best-fit total concentration BTCs versus the measured data [*Deng et al.*, 2004]. L denotes the travel distance. (c) The best-fit mobile concentration profiles versus the numerical snapshots [*Zhang et al.*, 2007]. (d) The best-fit groundwater age distributions at different wells versus TAD mobile concentration [*Weissmann et al.*, 2002]. See the text for details.

[*Schumer et al.*, 2003] significantly underestimates the tail decay rate for time $t > 1/\lambda \approx 1666$ yrs, and thus can misinform cleanup strategy at this site.

[17] The simulated groundwater age distributions (see symbols in Figure 2d) for different monitoring wells at Kings River alluvial fan (KRF) in California have been validated carefully using multiple environmental tracers [*Weissmann et al.*, 2002]. The (15 km-long, coarse-grain dominated) KRF alluvial depositional system contains multi-scale heterogeneities and does not follow a statistically homogeneous model. Consequently, the best-fit λ for the dimensionless probability density C of groundwater age in the mobile TAD model (8) differs between wells: $\lambda = 0.0075$ yr^{-1} for well 2-1 [$v = 110$ m/yr, $B = 180$ m^2/yr, $\gamma = 0.55$, $\beta_0 = 25.2$, $m_0 = 1$], $\lambda = 0.015$ for well 4-1 [$v = 60$, $B = 800$, $\gamma = 0.55$, $\beta_0 = 1.82$, $m_0 = 1$], and $\lambda = 0.018$ for well 5-1 [$v = 70$, $B = 120$, $\gamma = 0.55$, $\beta_0 = 6.04$, $m_0 = 1$].

[18] The exponential tempering parameter λ is related to retention time in immobile zones. *Zhang et al.* [2007] argue that the thickest immobile materials with the slowest decay rates control the transition of the late-time BTC from power-law to exponential, implying that λ codes the retention time in the largest immobile blocks. For a non-stationary heterogeneous medium (such as KRF), the heterogeneity structure varies, so that λ becomes space dependent. A future study will explore the quantitative relationship between system heterogeneity and particle waiting times, and the extension of the TAD model to capture the spatio-temporal nonstationarity.

4. Conclusions

[19] A novel tempered anomalous diffusion (TAD) model is proposed to capture the pre-asymptotic behavior of passive tracers in heterogeneous aquifers. The model imposes an exponential cutoff to power-law waiting times in the immobile zone, according to an adjustable truncation parameter. As time evolves, the TAD model transitions from time-fractional advection dispersion to classical (asymptotic) advection dispersion behavior. Exact analytical solutions and particle tracking methods are presented. Several examples demonstrate the applicability of the model for capturing the complete transport process for passive tracers through various systems at various scales. Natural media with complex solute mass exchange mechanisms (such as Case 2) or strongly heterogeneous media dominated by immobile phases (such as Case 3) can have relatively strong persistence of anomalous diffusion, causing slow transition to the asymptotic limit. In all examples, although the convergence to advection dispersion behavior is slow, the time-fractional advection dispersion model of *Schumer et al.* [2003] typically over-predicts late-time plume concentration. Similarly, the asymptotic advection dispersion model under-predicts late-time concentrations. Both kinds of prediction errors can seriously misinform contamination cleanup strategy and evaluation. The TAD model provides a simple but effective alternative that interpolates between these two end-members. It provides an accurate prediction of late-time concentrations and plume profiles in all cases examined, over a wide range of scales.

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