

On characterization of anomalous dispersion in porous and fractured media

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Abstract. A key characterization of dispersion in aquifers and other porous media has been to map the effects of inhomogeneous velocity fields onto a Fickian dispersion term (D) within the context of the conventional advection-dispersion equation (ADE). Recent compilations of data have revealed, however, that the effective D coefficient is not constant but varies systematically with the length or timescale over which transport occurs. A natural strategy to encompass this "anomalous" behavior into the context of the conventional ADE is to make D time dependent. This approach, to use $D(t)$ to handle the same anomalous dispersion phenomena, has also been common in the field of electronic transport in disordered materials. In this paper we discuss the intrinsic inadequacy of considering a time-dependent dispersivity in the conventional ADE context, and show that the $D = D(t)$ generalization leads to quantifiably incorrect solutions. In the course of proving this result we discuss the nature of anomalous dispersion and provide physical insight into this important problem in hydrogeology via analysis of a class of kinetic approaches. Particular emphasis is placed on the effects of a distribution of solute "delay times" with a diverging mean time, which we relate to configurations of preferential pathways in heterogeneous media.

Introduction

In recent years it has become evident that dispersion of solutes migrating through porous and fractured media behaves, in many cases, in a non-Fickian manner, with dispersion depending on the length or timescale over which the transport process occurs [e.g., *Lallemant-Barres and Peaudecerf*, 1978; *Gelhar et al.*, 1985, 1992; *Hewett*, 1986; *Abelin et al.*, 1988]. The usual explanation [e.g., *Neuman*, 1990] for this phenomenon is that as a solute migrates, it encounters heterogeneities at various scales, which lead to systematic deviation from average parameter values. Although this type of behavior is becoming more familiar in the study of heterogeneous and disordered systems, it is such a strong departure from our usual ideas concerning the use of averaged, intrinsic properties of a medium that one can denote it as "anomalous dispersion."

Traditionally, porous and fractured media have been modeled on the basis of a continuum approach, which is predicated on the ability to define a suitable "representative elementary volume" (REV) for the considered domain. Assuming the applicability of the REV approach, the resulting macroscopic equation that has been derived (in several ways) to describe miscible solute transport is the well-known advection-dispersion equation (ADE) [e.g., *Bear*, 1972]. The development of the term describing dispersion inevitably involves the fundamental assumption that macroscopic dispersion is macroscopically diffusive (Fickian). As such, macroscopic dispersion is assumed to be a constant quantity, dependent only on the mean velocity field and on dispersivity parameters that are considered to be intrinsic to the porous medium.

However, in light of the non-Fickian nature of dispersion, REV approaches are becoming increasingly eschewed in favor

of more general stochastic approaches. As a consequence a large number of works have used stochastic approaches to treat geological formations as statistically heterogeneous over multiple scales, and to derive appropriate transport equations that are spatially and/or temporally nonlocal [e.g., *Cushman and Ginn*, 1993; *Glimm et al.*, 1993; *Neuman*, 1993; *Beckie et al.*, 1994]. Solute transport can also be examined with simulators in structured media by solving for deterministic geometries (such as layered systems or domains with specifically defined heterogeneities [e.g., *de Marsily*, 1986]) or by using a statistical definition of the structure (such as percolation and fractal-based constructions [e.g., *Berkowitz and Balberg*, 1993; *Grindrod and Impey*, 1993]).

The theory of transport in random media, of which porous media constitute one example, spans a large literature. In an historical perspective the nonlocal transport phenomena being studied in the stochastic framework have been examined in other (parallel) fields for many years. For example, a variety of approaches are based on a Green's function, G , the propagator of a solute particle from an initial position to another one, in a specific configuration. An equation can then be developed for the configuration-averaged Green's function $\langle G \rangle$; these approaches all lead to similar results that $\langle G \rangle$ obeys a nonlocal equation. This equation can be specialized, by a Taylor expansion, to a generalized diffusion equation of the type derived by, for example, *Cushman and Ginn* [1993] (see, e.g., *Haan and Zwanzig* [1978], *Gochanour et al.* [1979], and *Klafter and Silbey* [1980]).

In addition to appearing in heterogeneous aquifers and petroleum reservoirs, the phenomenon of anomalous dispersion has now been well-documented in a range of problems involving electron and/or microscopic defect motion in disordered solids [e.g., *Scher et al.*, 1991]. All of these phenomena have demonstrated the need to invoke an anomalous dispersion of a transporting species. The apparent common feature in all of

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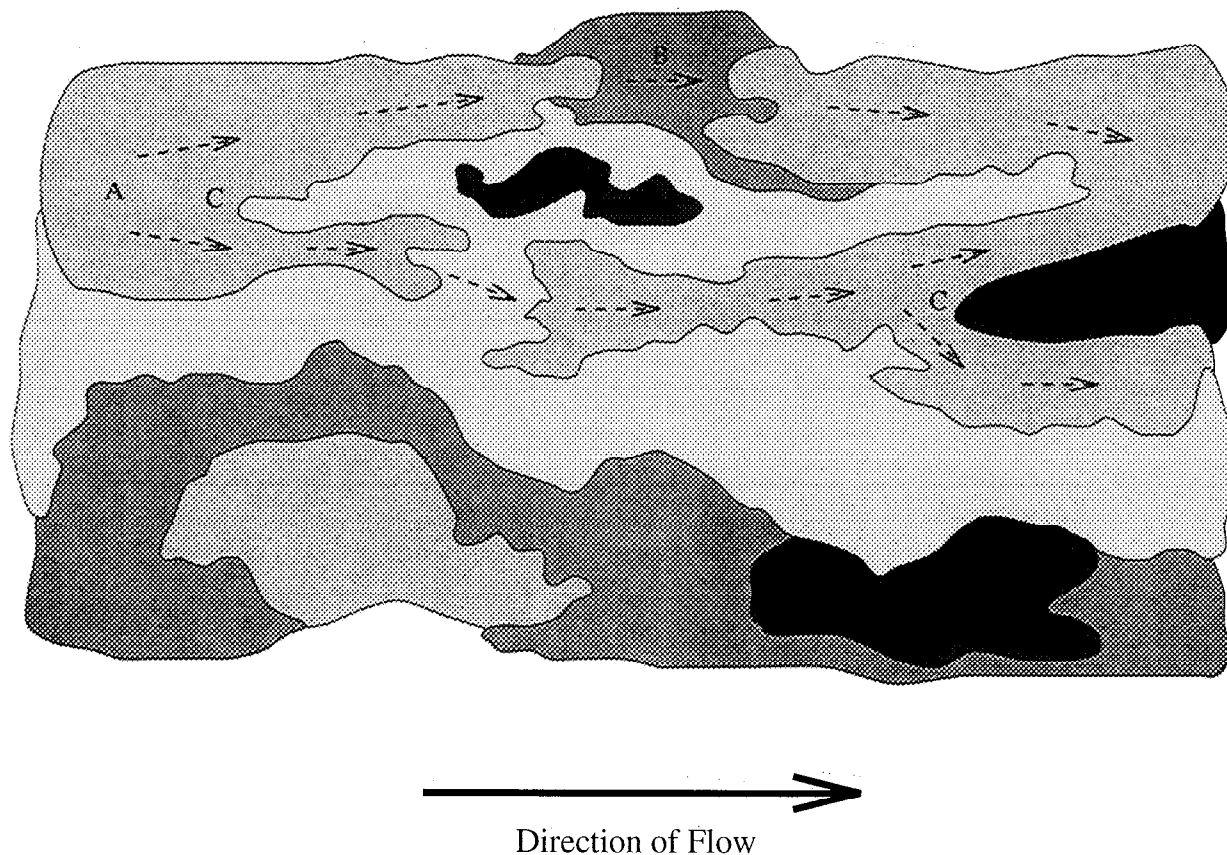


Figure 1. Schematic illustration of preferential pathways (channels) of flow in highly heterogeneous media (after Plate 2 of *Grindrod and Impey* [1993]); the shaded regions correspond to different values of velocity. Solute migration in the channel (A) can be delayed by a transition to a lower velocity (B) and/or by channel division (C). These changes act as “traps” to the channel flow.

these systems is the presence of a sufficiently random transport medium, which gives rise to a mean square displacement $\langle r^2 \rangle$ varying as t^ϕ , where ϕ generally does not equal unity as in Fickian diffusion (see, e.g., *Berkowitz and Braester* [1991] and *Neuman* [1990] for further discussion).

In spite of the large number of stochastic and scaling theories currently available, their perceived complexity and formalism have permitted the conventional ADE framework to remain popular, especially among practicing hydrogeologists. Moreover, references continue to appear suggesting that one can “set a time dependent dispersivity in the flow equation” [*Glimm et al.*, 1993, p. 115]. Within the framework of conventional approaches to describing solute transport, a phenomenological accounting in the literature for the distinctly non-Fickian behavior has been to introduce an effective time-dependent dispersion coefficient $\langle r^2 \rangle / t \equiv D(t) \sim t^{\phi-1}$ into the ADE and/or kinetic equations [e.g., *Vardeny et al.*, 1980; *Kakalios et al.*, 1987; *Glimm et al.*, 1993; *Grindrod and Impey*, 1993]. The purpose of this paper is to more fully discuss anomalous dispersion in the context of the conventional ADE, to illustrate the temporally nonlocal nature of this dispersion, and to show as a consequence that introducing a time-dependent dispersivity $D = D(t)$ leads to intrinsically and quantifiably incorrect results precisely under the conditions required for anomalous dispersion. In the course of the discussion we emphasize the significance of a distribution of solute “delay times” with a diverging mean time. An illustrative treatment of

the transport will relate these delays to configurations of preferential pathways in heterogeneous media.

Throughout this work we demonstrate the parallels of transport phenomena between solute migration in porous and fractured media and electron movement in disordered solids. Given the historical fact that many analyses of transport phenomena first appeared in the statistical physics literature, and in order to introduce aspects of the approaches, we retain some terminology from the physics literature in this paper, along with parallel concepts more familiar to the reader in hydrology.

The key physical idea is that anomalous dispersion is typically due to a wide distribution of delay times limiting the transport or flow in the medium. For purely advective flows these delay times are illustrated in Figure 1 and will be discussed in the final section. At any specific time t there are contributions to the transport from the range of delay times $\{\tau_i\}$, where $\tau_i < t$. We will show here that within the framework of a single equation to describe the motion of the dispersing species, the motion cannot be represented by an equation only containing terms evaluated at the same time t , that is, local in time, which would be the case if one used the function $D(t)$. Instead one must take account of the whole time history of the motion and thereby use a nonlocal (in time) equation. In other words, the price one has to pay to reduce all the coupled kinetics of the time-delaying processes into a single transport equation is nonlocality. In contrast to most stochastic ap-

proaches this mapping onto a nonlocal equation will be accomplished without configuration (or ensemble) averaging of a random variable. This serves our goal of presenting a clear physical origin of nonlocality in time. Afterwards, we discuss an important generalization of this intrinsic coupled-kinetic approach which is not limited to a spectrum of delay times $\{\tau_i\}$. We then show the limitation of using $D(t)$ within this framework. The essential ideas will be related to advective flows in a random medium in the discussion section. The full, formal development of our new approach will be reserved for another work. A preliminary account, applied to fractured aquifers, can be found in work by *Berkowitz and Scher* [1994].

We now first describe the physical process of a motion limited by extensive, multiple trapping (MT) [*Schmidlin*, 1977; *Muller-Horsche et al.*, 1987]. To orient the reader, the MT process is analogous to a simplified version of solute transport in a porous medium, wherein particles can enter regions of slow flow which can delay migration (i.e., channeling effects; see discussion section, below) and/or can be retarded in their advance by first-order nonequilibrium adsorption kinetics. Figure 1 is a schematic illustration of the model. We will later use this model to develop the conditions under which anomalous dispersion can appear. These conditions are subtle, and a nonlocal term per se is not sufficient. Apropos the remarks above, we do not contend that the MT model is the only explanation for the occurrence of temporal nonlocality. Rather, we present the MT model as a means to illustrate how the nonlocal representation can arise and to demonstrate simply how channeling effects in heterogeneous geological formations cannot be correctly described by stochastic approaches using limited permeability correlation lengths.

Mathematical Development

The MT process can be easily visualized as a particle flux with velocity \mathbf{v} interacting with states (or regions) in which the particles are temporarily delayed or immobilized. In the context of photoconduction, for which the MT process was originally developed, the mathematical formulation is a set of coupled kinetic equations for the particle densities p_i for the i th state or trap and p the density of the flowing particles [*Schmidlin*, 1977; *Muller-Horsche et al.*, 1987],

$$\partial p / \partial t = g(x, t) + \sum_i p_i \xi_i - p \sum_i \omega_i - \nabla \cdot \mathbf{f}_p \tag{1}$$

$$\partial p_i / \partial t = p \omega_i - p_i \xi_i \tag{2}$$

$$\mathbf{f}_p \equiv p \mathbf{v} - D \nabla p \tag{3}$$

Each trap is completely characterized by the rate of capture (ω_i) into and rate of release (ξ_i) from the trap; \mathbf{f}_p is the mobile particle flux with velocity \mathbf{v} , D is the regular diffusion coefficient, and $g(x, t)$ is the source term. The set of equations (1)–(3) completely defines the MT problem. In the context of solute transport in porous media, it is obvious that these equations also represent advective-dispersive transport in the presence of first-order kinetics, such as solute transport between mobile and immobile fluid regions [e.g., *Beven and Germann*, 1981; *van Genuchten*, 1981] or first-order kinetic nonequilibrium adsorption [e.g., *Cameron and Klute*, 1977].

We can solve the equations in Laplace space for p_i in terms of p and, substituting the relation into (1), derive one closed equation only involving p ,

$$\partial p(x, t) / \partial t = \int_0^t \phi(t - \tau) [g(x, \tau) - \nabla \cdot \mathbf{f}_p(x, \tau)] d\tau \tag{4}$$

where the Laplace transform $\bar{\phi}(u)$ of $\phi(t)$ is

$$\bar{\phi}(u) = 1 / [1 + \sum_i \omega_i / (u + \xi_i)] \tag{5}$$

The equation (4) for $p(x, t)$ is nonlocal in time; the $\phi(\tau)$ contains all the information pertaining to the distributed release of the mobile particles of previously delayed particles, $\tau < t$. The physical picture is clear; if one insists on using a local continuity equation for p , (1), one must explicitly include the local kinetics of the trapped particles. The MT process can give rise to anomalous dispersion; it depends on the relationship between the frequency of occurrence and the release rates of the trapping states, which will be fully discussed in the concluding section. As the structure of (4) and (5) demonstrates, this behavior cannot be affected by, for example, replacing the time history due to the terms which couple with the trapping states, by a local advective term and a Fickian term containing $D(t)$. This is the root cause of the difficulty of the use of $D(t)$ and below we will demonstrate this difficulty by comparison of the explicit solutions, determined by the two approaches, of the same physical process. We will further compare these solutions to the results of numerical simulation of the same problem. Note here a sharp departure from the usual stochastic approaches; the nonlocal equation (4) was derived without an ensemble average (the randomness being due to the spatial distribution of the traps).

With the understanding gained from the MT model we are naturally led to a more general approach to these problems using a random walk, where the discrete step number is replaced by the continuous time variable in the walk. For brevity we will refer to this continuous time random walk as CTRW. *Schmidlin* [1977] has shown that properly interpreted, (4) is equivalent to a special case of a CTRW. The purpose of introducing the CTRW process into our discussion here is that we can clearly indicate the difference in results using the CTRW and $D(t)$ approaches to the same problem. Moreover, important examples of anomalous dispersion in disordered solids, calculated with the CTRW formalism, have been thoroughly confirmed experimentally [e.g., *Scher et al.*, 1991].

The CTRW is a simple generalization of an ordinary random walk (where the continuation of the walk occurs at discrete time steps) on a lattice,

$$R(\mathbf{s}, t) = \sum_{\mathbf{s}'} \int_0^t \psi(\mathbf{s} - \mathbf{s}', t - \tau) R(\mathbf{s}', \tau) d\tau \tag{6}$$

where $R(\mathbf{s}, t)$ is the probability for a particle to just arrive at a site \mathbf{s} at time t , and $\psi(\mathbf{s}, t)$ is the probability per unit time for a transition between sites separated by \mathbf{s} and arrival times t . The proper choice of the ψ function is the main task in representing the stochastic process of interest. Note that (6) is now also nonlocal in space as well as in time. For electrons and microscopic defects moving in disordered solids it has been shown [*Scher et al.*, 1991, and references therein] that the ψ function can be represented by a power law decay $\psi \sim t^{-1-\beta}$ where $\beta > 0$ (only for $\beta \leq 1$ does one have completely anomalous behavior), and the β parameter is determined by the variables defining the specific transport model. Using this form of ψ in the CTRW, (6), for the case of biased transport (i.e., in

the presence of an applied finite mean velocity or an electric field), produces [Shlesinger, 1974] a highly dispersed pulse of initially injected particles with a variance $\sigma^2 = \langle r^2 \rangle - \langle r \rangle^2 \sim t^{2\beta}$ (for $\langle r \rangle \neq 0$). Note that 2β is equal to the ϕ exponent as discussed in the Introduction. Recent laboratory measurements of electron transport in polymer films have shown [Schein *et al.*, 1993] that this behavior persists up to the limit $\beta \rightarrow 1$ in the case of biased transport.

Proof of the Inadequacy of $D(t)$

We now consider a physical process where the total number of particles $N(t)$ undergoing this highly dispersive transport encounters absorbing sites (permanent traps) so that $N(t)$ decreases slowly in time. The decay of $N(t)$ in the presence of these purely absorbing sites is a direct measure of the underlying effective anomalous dispersion properties of the transport. Analytical results using CTRW, for $N(t)$, with the absorbing sites in a plane perpendicular to the applied bias [Scher and Montroll, 1975], yield

$$N(t) \sim t^{-\beta} \quad (7)$$

The same results were obtained for absorbing sites distributed periodically (with arbitrary period) [Scher, 1981]. For randomly distributed absorbing sites we have to resort to simulation studies. The same behavior as in (7) was found again for both bias and no bias and in any spatial dimension [Klafter *et al.*, 1986]. Thus the solution to the “trapping” problem for the number of “survivors” undergoing dispersive transport or diffusion is the power law t -dependence in (7).

The diffusion case (i.e., no bias) of the same problem is treated phenomenologically in the literature by using a $D(t) = \langle r^2 \rangle / t \sim t^{\beta-1}$ (since for the unbiased case, $\psi \sim t^{-1-\beta}$ gives rise to a $\langle r^2 \rangle \sim t^\beta$) and a trapping rate (into the absorbing sites) $R \propto D(t)$ [Vardeny *et al.*, 1980; Kakalios *et al.*, 1987], such that

$$dN(t)/dt = -RN(t) \propto -t^{\beta-1}N(t) \quad (8)$$

One can easily integrate (8) to obtain

$$N(t) \sim \exp(-ct^\beta) \quad (9)$$

which is the so-called “stretched exponential” function (where c is a coefficient of proportionality). The solution to the trapping problem in (9) is in sharp contrast to the correct one in (7). Hence the use of $D(t)$ leads to untenable physical results by sharply diminishing the effects of the dispersion (i.e., by de-emphasizing the slower part of the distribution). We note also that the “stretched exponential” behavior of $N(t)$ can be generated by a different process: the “target” problem. We refer the interested reader to the work of Shlesinger and Montroll [1984] for a full discussion of this problem, which has been successfully used to describe diverse relaxation phenomena in disordered media, such as mechanical relaxation after the removal of an applied load.

Discussion

The inability to “patch” into the conventional ADE an empirical $D(t)$ to represent scale-dependent dispersion represents a serious challenge to the use of this equation, which is derived by spatial averaging (REV) approaches. One must consider whether such approaches are appropriate for these

kinds of aquifers or, more generally, the type of equations that should be used as a new starting point. As mentioned in the Introduction, stochastic approaches offer a general framework for describing solute transport, although there exist inherent limitations in some of their applications [e.g., Dagan, 1989; Gelhar, 1993; Tanksley and Koplík, 1994]. In this concluding section we will consider complementary approaches and use one of them to provide more physical insight into the subtle conditions needed for anomalous behavior. This subtlety can be seen with the following familiar examples.

Consider a fracture network with a range of fracture sizes and apertures. If the solute and water flow through a subnetwork of fractures and most of the flow is then funneled into a fracture much larger than any in the subnetwork, the effect of the large fracture is simply to “translate” the dispersed solute, that is, to change the timescale of the flow but not the dispersion. In the opposite case where most of the flow can avoid the large fracture then there is a minor, leading edge “blip” to the dispersion and no effect on the timescale of the flow. Thus the mere presence of even a large range in fracture sizes is not enough to produce scale-dependent dispersion or rule out the use of a suitable REV. It is the connectedness or “encounter” of the flow with the range of fracture sizes that is also crucial. The encounter-range relationship has to be such as to produce a wide spread of interacting flow paths each with a different sequence or different set of fracture size encounters. If after a sufficiently long time each path has the same average set, then the dispersion will approach a constant value. These issues are also directly relevant to heterogeneous porous media. Many natural formations display significant heterogeneity (at least in terms of the flow field), and the existence of preferential flow pathways and channels in such media is now well established [e.g., Winograd and Pearson, 1976; Neuman, 1990; Grindrod and Impey, 1993; Jussel *et al.*, 1994; Moreno and Tsang, 1994].

Further discussion of the encounter-range relationship is best illustrated in the context of a model that can also serve as a possible starting point other than the conventional ADE. We consider the model shown schematically in Figure 1 and in (1)–(3). At any point in the “free flow” the solute, moving along a preferred flow pathway or channel, can make a transition to a region of slow flow or be diverted and delayed until it manages to return; Grindrod and Impey [1993] employ a similar approach with their channel network picture. In this model we have to specify the range of velocities v and the relative density of regions with v , $\rho(v)$. It should be emphasized that ρ is not the distribution of the entire velocity field in the aquifer but is determined from those structural features of the aquifer that cause delays. We assume each delay time varies inversely with v , where both v and $\rho(v)$ are normalized to an appropriate reference velocity and the density at that same velocity, respectively.

Now in addition to the range, as discussed above, we have to consider the encounter, which should simply be proportional to $\rho(v)$ in a model with uniform access to each region. We assume that in general, the flow path of a solute in a strongly heterogeneous formation encounters many short delays and very rarely a large one. However the rare large one can have a significant impact on the dispersion which, reiterating, is the prime reason for scale-dependent dispersion within this framework. The density should then drop off as $v \rightarrow 0$ and we can consider the basic case of a power law decay

$$\rho(v) \sim v^\beta \quad (10)$$

This expression can be derived from the most prevalent example of MT in electron transport in amorphous semiconductors [Tiedje, 1984] and polymeric films [Muller-Horsche et al., 1987].

It can be shown from the analogous electronic transport [Scher et al., 1991, and references therein] that the exponent β in (10) is the same as in, for example, (7), and further that the criterion for anomalous dispersion derived from (10) is the same: for $\beta < 1$ the dispersion is anomalous while for $\beta > 1$ it is not. Thus for the same available range of v for the slow flow regions, one can have either type of dispersion depending on the relative encounter of the flowing solute with these regions. The power law form for ρ enables a simple adjustment of this relative fall-off in v . Note that a lognormal functional dependence of ρ with v , for example, has a uniformly faster decay than a power law so that after an initial phase, the dispersion will rapidly become scale independent.

As discussed in the Introduction, the complexities of inhomogeneous velocity fields in aquifers have spurred the use of stochastic methods [e.g., Dagan, 1989; Gelhar, 1993] and simulations of flow in a variety of assumed structures. In addition to the study of nonlocal phenomena the existence and properties of convergence to a constant "macrodispersion" coefficient have been analyzed extensively [e.g., Dagan, 1988]. As frequently noted, however, most of the theoretical studies within the stochastic framework assume relatively small variations in the hydraulic conductivity field. Thus, we emphasize that it is precisely the critical (rare) long delay events discussed above that are neglected (e.g., higher order terms in perturbation expansions) in the homogenization techniques that are implicitly or explicitly incorporated into many stochastic (as well as deterministic) derivations. Quantitative limitations to the applicability of these methods to heterogeneous flow fields are given, for example, by Glimm et al. [1993] and Tanksley and Koplík [1994]. We also note that the above arguments regarding the encounter-range relationship, and the nature and effects of the critical long delay events, are independent of scale and can account for geological heterogeneity over multiple scales (such as those discussed by Neuman [1990]). A physically plausible approach which analyzes the key correlations in the flow pattern [e.g., Grindrod and Impey, 1993] and develops the formalism to accommodate them (e.g., Berkowitz and Scher [1994], applying the CTRW formalism to fractured aquifers) would seem to hold considerable promise.

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