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Diffusion in one-dimensional multifractal porous media

S. Lovejoy
Department of Physics, McGill University, Montreal, Quebec, Canada

D. Schertzer
LMM (CNRS) Boite 162, Université P. & M. Curie, Paris, France

P. Silas
Department of Physics, McGill University, Montreal, Quebec, Canada

Abstract. We examine the scaling properties of one-dimensional random walks on media with multifractal diffusivities, which is a simple model for transport in scaling porous media. We find both theoretically and numerically that the anomalous scaling exponent of the walk is $d_w = 2 + K(-1)$ where $K(-1)$ is the scaling exponent of the reciprocal spatially averaged ("dressed") resistance to diffusion. Since $K(-1) > 0$, the walk is subdiffusive; the walkers are effectively trapped in a hierarchy of barriers. The trapping is dominated by contributions from a specific order of singularity associated with a phase transition between anomalous and normal diffusion. We discuss the implications for transport in porous media.

1. Introduction

Diffusion is the simplest transport mechanism of interest in porous materials and more generally in random media; it has been extensively studied (see, e.g., reviews by *Havlin and Ben-Avraham* [1987] and *Bouchaud and Georges* [1990]). Since many natural (especially geophysical) phenomena are scaling, it is natural to consider diffusion in scaling media; indeed, many studies have been made of percolating and hierarchical systems. In the latter systems there are long-range correlations; the diffusion is confined to a fractal set, the media are monofractal, and their scaling is characterized by a unique fundamental exponent. In porous media, geometric fractal sets have been used as binary models for hydraulic conductivity [*Wheatcraft et al*., 1990], as has monofractal fractional Brownian motion [*Molz and Boman*, 1993; *Molz and Liu*, 1997]. Similarly, *Gabriel et al*. [1986], *Lovejoy and Schertzer* [1989], *Cahalan and Joseph* [1989], *Lovejoy et al*. [1990], *Gabriel et al*. [1990], and *Davis et al*. [1990] have used fractal sets as models of clouds and investigated the (anomalous) radiative transport. Since the radiative transfer (or kinetic, Boltzman) equation can also be used for modelling transport in porous media, the latter results are also relevant to porous media.

It is increasingly clear that scaling dynamical processes (such as those associated with turbulent cascades) will be multifractal rather than monofractal, generally requiring an infinite hierarchy of exponents for its specification. Over a dozen geophysical fields have been reported to be multifractal over various ranges (see recent reviews by *Schertzer and Lovejoy* [1998] and *Lovejoy and Schertzer* [1995]), and *Liu and Molz* [1997] have recently reported empirical analyses of the logarithm of hydraulic conductivities concluding that the latter were indeed consistent with universal multifractals estimating the three fundamental parameters.

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The primary consequence of the long-range correlations implicit in the scaling is that the diffusion has an anomalous exponent (i.e., the variance of the distance traveled by a diffusing particle is a nonlinear power law in time). It is therefore surprising that in spite of the obvious theoretical and empirical interest of multifractals that very little attention has been paid to the corresponding transport properties. (The long-range correlations in multifractals are sufficiently strong so that they are outside the scope of analytic stochastic methods such as those of *Marle et al*. [1967], *Gelhar et al*. [1979], and *Matheron and de Marsily* [1980]; see Appendix A for more details). The main exception is the study of radiative/kinetic transport in multifractal media where some early results have been obtained [e.g., *Davis et al*., 1993; *Lovejoy et al*., 1995; *Naud et al*., 1996]. As for the simpler transport problem of diffusion in multifractal media, there is practically nothing in the literature; the primary reference is by *Meakin* [1987], who examined the properties of random walks on rather special ("microcanonical") multifractals generated by discrete cascades in two spatial dimensions. An important limitation, underlined by *Marguerite et al.* [1998], of this essentially numerical study was that it considered the rms distance of the walkers as functions of the number of steps, not of the diffusion time; his study was of random walks not diffusion per se. Another relevant exception was work by *Weissman* [1988], who discussed certain non-normalized multiplicative processes, without averaging over initial particle positions. A final related paper is one by *Saucier* [1992], who studied the effective transport properties of certain multifractal permeability fields using renormalization group methods.

An early progress report on this work is given by *Silas et al*. [1993], and more details can be found in work by *Silas* [1994]; see also work by *Lovejoy and Schertzer* [1995]. For (qualitatively different) numerical results and phenomenological arguments in two spatial dimensions see work by *Marguerite et al*. [1998]. We treat the seemingly general case of diffusivities with convergent harmonic averages (finite $K(-1)$; see below), and we test the theory with numerical simulations of lognormal multifractals. (The term "lognormal multifractal" is actually a bit of a misnomer since only the low-order statistics are lognormal; see Appendix A for the divergence of moments). However, there are theoretical (I. Tchiguirinskaia and F. Molz, manuscript in preparation, 1998) and empirical reasons [*Liu and Molz*, 1997] to support the possibility that the realistic diffusivities are nonanalytic and that $K(-1)$ does not converge (the precise conditions for the validity of our results are discussed below). Thus this restriction, combined with the special (and unrealistic) nature of one-dimensional diffusion implies that these results on the simplest multifractal transport problem are primarily of theoretical interest.

2. Multifractal Resistivity Fields

Consider a stochastic one-dimensional multifractal density field (e.g., generated by a multiplicative cascade process; see Figure 1) denoted $\rho_\lambda(x)$ where $\lambda > 1$ is the ratio of the largest scale of interest to the smallest scale of homogeneity displaying singularities γ:

$$\rho_\lambda \sim \lambda^\gamma \tag{1}$$

satisfying a well defined probability distribution at each scale:

$$\Pr(\rho_\lambda > \lambda^\gamma) \approx \lambda^{-c(\gamma)} \tag{2}$$

"Pr" indicates "probability," $c(\gamma) \geq 0$ is the codimension function, and equality is to within slowly varying (e.g., logarithmic) prefactors. Equation (1) should be considered as a change in variable; at each point $\gamma = \log \rho_\lambda / \log \lambda$; (2) then describes how the histograms/probability distributions of γ vary as functions of scale λ. As the resolution (λ) increases, the values of ρ corresponding to a given γ (>0) grow larger while simultaneously becoming more rare; that is, the field at each singularity level becomes sparse (as quantified by $c(\gamma)$). The statistical moments of the multifractal field are then described by the moment scaling function $K(q)$:

$$\langle \rho_\lambda^q \rangle = \lambda^{K(q)} \tag{3}$$

where q is the order of the moment, and the brackets indicate ensemble averaging. The exponents $K(q)$, $c(\gamma)$ are related through a Legendre transformation which establishes a one-to-one correspondence between moments and singularities: $q = c'(\gamma)$, $\gamma = K'(q)$.

For those familiar with the more popular dimension ($f(\alpha)$) formalism [*Halsey et al.*, 1986], it is worth mentioning the relationship between the dimension and codimension formalism adopted here. In a finite dimensional (d) observing space the relationship is $\tau_d(q) = d(q-1) - K(q)$, $f_d(\alpha_d) = d - c(\gamma)$, $\alpha_d = d - \gamma$. The subscripts have been added to emphasize their dependence on the dimension of space. Since we consider stochastic processes with an infinite dimensional probability space (i.e., $d \to \infty$), we use the codimension multifractal formalism [*Schertzer and Lovejoy*, 1987, 1992] which always has exponents independent of d.

The coefficient of diffusion $D_\lambda(x)$ (in porous media, depending on the problem, either the diffusivity or the hydraulic conductivity; see below) is here taken to be

$$D_\lambda(x) = \frac{1}{\rho_\lambda(x)} \tag{4}$$

that is, ρ_λ is a resistance to diffusion; regions of high resistance to diffusion are likely to correspond to rare, dense (impenetrable) regions of the medium. The question of whether to consider ρ or its inverse to be the conserved multifractal quantity is a question of physics: Is it more realistic to assume that the rare, sparse regions correspond to "bottlenecks" (resistances) or, conversely, to regions of rapid transport (large diffusion coefficient)? See Appendix B for extensions to the case where the conservative multifractal process is the diffusive conductivity rather than resistivity; this is indeed *Liu and Molz*'s [1997] interpretation of their empirical borehole porosity data.

In the following we consider the time-dependent diffusion equation

$$\nabla \cdot (D(\mathbf{x}) \nabla P) = \frac{\partial P}{\partial t} \tag{5a}$$

where P is the diffusing quantity. In hydrogeology, the diffusion equation arises as a model both for transport of solute and for flow of groundwater. For solutes the time-dependent diffusion equation (5a) arises with D as the diffusivity and P as the solute concentration in the limit where the advection velocity $\mathbf{v}$ is negligeable. In groundwater transport the steady state diffusion equation arises when Darcy's law holds: $\mathbf{v} = D\nabla P$ and the fluid is incompressible ($\nabla \cdot \mathbf{v} = 0$), that is, if D is the hydraulic conductivity and P is the pressure. (In the one-dimensional case treated below, the incompressibility condition implies constant $\mathbf{v}$; the corresponding one-dimensional solute diffusion with multifractal diffusivity can then be derived from the pure ($\mathbf{v} = 0$) diffusion equation (5a) by a simple change of variables). Other related transport problems where diffusive transport on multifractals may be relevant include radiative transfer since the diffusion approximation to radiative transfer is obtained by taking $\rho_\lambda(x)$ proportional to the (multifractal) optical density in the limit of large ρ. (Note that to model clouds, the conservative multifractals discussed here must be given a fractional integration (i.e., a power law fourier space filter) of order $\approx$1/3; see work by *Schertzer and Lovejoy* [1987] and *Schertzer et al.* [1997]). Another possible geophysical application is for the diffusion of heat in rock with multifractal thermal diffusion coefficient D (with or without heat sources); in this case P is the temperature. In still other systems, $\rho_\lambda(x)$ could be identified with the electrical resistivity, inverse permeability, etc.

The simplest nontrivial multifractal diffusion problem is the one-dimensional time-dependent diffusion equation obtained by taking $\nabla \to \partial/\partial x$ in the above. Although we will give a simple analytic argument for describing this one-dimensional diffusion, it will be important to check the results with numerical simulations. Such simulations may be performed with many different numerical techniques; probably the simplest (and also the most widely used in statistical physics) is the Monte Carlo random walk technique, which recognizes (5a) as the Fokker-Plank equation for the probability density (P) of a (drift-free) Langevin equation (the continuous space limit of a random walk). In this case numerical techniques are used to solve the "master equation" (the continuous time, discrete space diffusion equation). The large-scale properties (where the spatial discretization is unimportant) then approximate those of (5a). Specifically, consider a single random walk performed on a single realization of the multifractal constructed on the unit interval over a large-scale ratio Λ (i.e., the smallest scale is Λ^{-1}) and which obeys the master equation:

$$\frac{dP_\Lambda(x_i, t)}{dt} = T_\Lambda(x_{i+1} \to x_i) P_\Lambda(x_{i+1}, t) - T_\Lambda(x_i \to x_{i+1}) P_\Lambda(x_i, t) + T_\Lambda(x_{i-1} \to x_i) P_\Lambda(x_{i-1}, t) - T_\Lambda(x_i \to x_{i-1}) P_\Lambda(x_i, t) \quad (5b)$$

where P_Λ is the probability of finding a particle at the site x, time t. The consecutive sites x_i, x_{i+1} are separated by a distance Λ^{-1}. The T_Λ's are the transition rates; to approximate the diffusion equation the transition rates must be related to the site to site transmission coefficients $\sigma_\Lambda(x_i; x_{i\pm1})$ and diffusivities as follows [*Aziz and Settari*, 1979]:

$$T_\Lambda(x_i \to x_{i\pm1}) = \frac{\sigma_\Lambda(x_i; x_{i\pm1})}{\sigma_\Lambda(x_i; x_{i+1}) + \sigma_\Lambda(x_i; x_{i-1})}$$
$$\sigma_\Lambda(x_i; x_{i\pm1}) = \frac{2D(x_i)D(x_{i\pm1})}{D(x_i) + D(x_{i\pm1})} = \frac{2}{\rho_\Lambda(x_i) + \rho_\Lambda(x_{i\pm1})} \quad (6)$$

Note that in modeling the diffusion of solutes, the interpretation of the walkers is fairly straightforward: Each walker can be identified with a particle of solute, and the walker concentration is identified with the solute concentration. Although the corresponding groundwater equation is time independent, the random walk Monte Carlo technique can still be used as long as the boundary conditions are time independent and we seek the long-time walker concentrations. In this case the interpretation is less obvious: The walker concentration gives the pressure, the gradient of the concentration being proportional to the fluid velocity.

3. Diffusion Properties

In one spatial dimension with independent identically distributed (time-independent) random transition rates where $\langle\rho\rangle$ is finite, in the limit of long times and large distances, the properties of the walk can be derived by taking Fourier and Laplace transforms of (5a) (in space and time respectively; see work by *Machta* [1981] and *Zwanzig* [1982]). We now use a (nonobvious) extension of these long-time, large-distance results [*Havlin and Ben-Avraham*, 1987; *Weissman*, 1988]:

$$\rho_{\text{eff}} = \frac{1}{D_{\text{eff}}} = \frac{t}{\overline{x^2}} \approx \frac{1}{N}\sum_{i=1}^{N}\frac{1}{D_i} = \frac{1}{N}\sum_{i=1}^{N}\rho_i \quad (7)$$

where N is the number of distinct sites visited by the random walker and the ρ_i are the resistances associated with those sites, t is the time taken for the walk, and $\overline{x^2}$ is the variance of walks on a single realization of the (multifractal) process (overbars indicate means over walks, angle brackets indicate means over ensembles of multifractal realizations of $\rho(x)$). This states that the random walker experiences an effective resistance $\rho_{\text{eff}} = (D_{\text{eff}})^{-1}$ equal to the mean resistance of the sites it has visited.

Divide the multifractal into λ disjoint regions each with $N = \Lambda/\lambda \gg 1$ distinct sites (i.e., over lengths λ^{-1}). Equation (7) now yields the effective diffusion coefficient in the jth interval of length λ^{-1}:

$$\rho_{\lambda,j,\text{eff}} = D_{\lambda,j,\text{eff}}^{-1} = \frac{\lambda}{\Lambda}\sum_{i=1}^{\Lambda/\lambda}\rho_{\Lambda,j,i} \approx \rho_{\lambda,j,d} \quad (8)$$

where the sum is over all the $\rho_{\Lambda,i}$ for the $N = \Lambda/\lambda$ sites in the interval. The approximation $\rho_{\lambda,\text{eff}} = \rho_{\lambda,d}$ is valid if $\Lambda/\lambda \gg 1$, that is, when many sites are visited. The quantity $\rho_{\lambda,j,d}$ is the spatial average over scale λ of the multifractal ρ in the limit $\Lambda \to \infty$ (i.e., over a completed cascade). The subscript d denotes "dressed" and is necessary to distinguish the latter average of a completed process from the "bare" process developed only down to a resolution λ (i.e., without the small-scale interactions). The bare/dressed distinction [*Schertzer and Lovejoy*, 1987] is necessary in canonical multifractals and is fairly well understood. For singularities below a critical value $\gamma < \gamma_D$ (the case of interest here) the statistics of ρ_λ, $\rho_{\lambda,d}$ are the same to within unimportant factors of order 1 (i.e., the spatial averaging kills the high-frequency variability, leaving only the low-frequency component ρ_λ); for $\gamma \geq \gamma_D$, on the contrary, the small-scale variability becomes dominant, the dressed codimension becomes linear, and the corresponding moments diverge. Even for weak events (low γ), the approximation $\rho_\lambda, \approx \rho_{\lambda,d}$ is valid only as long as $c(\gamma) > 0$; hence it will not always be true in the universal multifractals with Levy index $\alpha < 2$ where $c(\gamma) = 0$ for all γ below a critical value. This may be relevant since *Liu and Molz* [1997] have performed empirical analysis of the logarithm of borehole hydraulic conductivities (see Appendix B) and found $C_1 \approx 0.05$ and Levy parameter α in the range 1.3–1.9.

If we consider only a single starting position, then for a conserved multifractal ($\langle\rho_\lambda\rangle$ = constant) (8) shows that we obtain the result of *Weissman* [1988], that is, no anomalous scaling. By considering the average over various initial walker starting positions, we sample different parts of the resistivity field; this is equivalent to ensemble averaging (see, however, the caveat about ergodicity below). It is precisely the effect of this averaging over D_λ in these stochastic multifractals which (as in work by *Meakin* [1987]) yields the anomalous result. Averaging over all N intervals of scale λ^{-1}, we obtain

$$\langle D_\lambda\rangle \propto \langle\rho_{\lambda,d}^{-1}\rangle \propto \lambda^{K_d(-1)} \propto \lambda^{K(-1)} \quad (9)$$

where we have used $K_d(-1) = K(-1)$, a relation which will hold only if $K(-1)$ is finite (hence it will not apply to the universal multifractals [*Schertzer and Lovejoy*, 1987; *Schertzer et al.*, 1995] with non-Gaussian, $\alpha < 2$ generators).

Using (7) and (9) and $L/\lambda = \langle x^2\rangle^{1/2}$ for the extent of the walk, we obtain

$$t \approx \frac{\langle\overline{x^2}\rangle}{\langle D_\lambda\rangle} \approx \langle\overline{x^2}\rangle^{1+(K(-1)/2)} \quad (10)$$

Note that in the above, we have ignored the uninteresting L-dependent factors and we have replaced the average over the λ disjoint intervals of the realization with the ensemble average indicated with the angle brackets. This is legitimate as long as the singularity giving the dominant contribution to $\langle D_\lambda\rangle$ is almost surely present in the sample, a condition which, because of the general nonergodicity of the process, is not automatically verified, but which will nevertheless hold here. (Following *Schertzer and Lovejoy* [1989], the maximum and minimum singularities present in a single realization ($\gamma_{\max}$, $\gamma_{\min}$) can be estimated from $c(\gamma_{\max}) = c(\gamma_{\min}) = D$; the corresponding maximum and minimum orders of moment are $q_{\min} = c'(\gamma_{\min})$ and $q_{\max} = c'(\gamma_{\max})$. For the lognormal case (see below) we obtain $\gamma_{\min}^{\max} = -C_1 \pm 2\sqrt{C_1 D}$, $q_{\min}^{\max} = \pm\sqrt{D/C_1}$. Therefore $q_{\min} < -1$ as long as $C_1 < D$, so that in this case the $q = -1$th order moment and hence the γ_{-1}

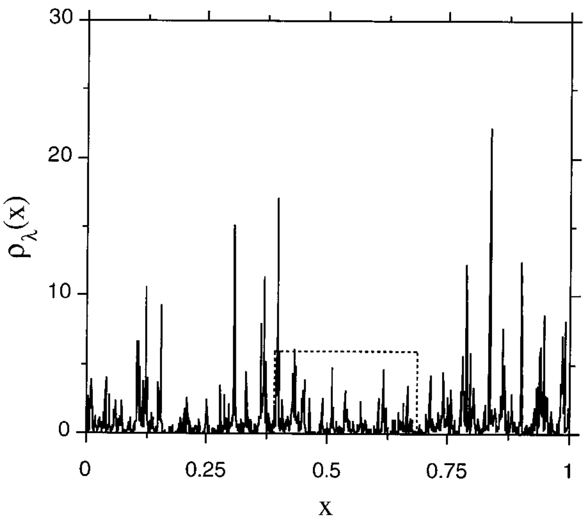

Figure 1. A one-dimensional lognormal multifractal field with $C_1 = 0.2$ and $\lambda = 1024$.

singularity is almost surely present for every realization of a lognormal multifractal. Since $C_1 < D$ is also the condition that the multifractal is nondegenerate, in practice this condition will always be satisfied in lognormal multifractals).

Defining the exponent d_w by $t \approx \langle x^2 \rangle^{d_w/2}$ and comparing with (10) we obtain

$$d_w = 2 + K(-1) \tag{11}$$

hence, since $K(-1) > 0$, $d_w > 2$, we will have subdiffusive behavior; as noted by *Meakin* [1987], the particles are trapped in a hierarchy of barriers. In the limit $\Lambda \to \infty$ the diffusive behavior is therefore totally dominated by structures with resistivity singularity $\gamma_{-1} = K'(-1)$ distributed over a fractal set with codimension $c(\gamma_{-1})$. The higher-order singularities are too rare to affect the transport, and the lower-order singularities are too weak to significantly trap the particles. This critical singularity is associated with a phase transition: If the resistivity field is replaced by a thresholded field with all values exceeding a fixed T reset to the value T, then in the limit $\Lambda \to \infty$ there will be a transition from anomalous diffusion (with the above exponent) to normal diffusion when T is reduced below the critical value $\Lambda^{\gamma_{-1}}$. Appendix C gives some relevant numerical confirmation of this dynamical phase transition for the lognormal multifractal.

Once again, the fundamental role of averaging over initial positions is brought out by comparison with the (deterministic) "hierarchical" models [e.g., *Havlin and Ben-Avraham*, 1987]. In the latter the particles start at a fixed origin, typically the center of the deterministic singularity in D, $\gamma_{D,d}$ (i.e., at a location where $D_{\lambda,d} \approx \lambda^{\gamma_{D,d}}$). Since there is no averaging over initial starting positions (the hierarchical models are deterministic and are not spatially homogeneous), we obtain $d_w = 2 + \gamma_{D,d}$. This shows the critical role played by the averaging over the starting positions (and hence over singularities γ) in the stochastic multifractals.

As a final comment, although the steady state probability density is multifractal, requiring in general an infinite number of exponents for its specification, the statistical moments $\langle \overline{x^q} \rangle$ scale with a single exponent (the overbar indicates an average over an ensemble of walkers on an individual realization), $\langle \overline{x^q} \rangle \approx t^{S(q)}$ where $S(q) = H_w q$, and $H_w = 1/d_w$ is the gap exponent (equal to 1/2 for normal diffusion). This monoscaling of the moments is presumably a consequence of the fact that a random walk is a random additive process whereas multiscaling arises from multiplicative processes.

4. Numerical Tests

We now test the above result numerically on lognormal universal multifractals characterized by $K(q) = C_1 q(q - 1)$, $c(\gamma) = (C_1 + \gamma)^2/(4C_1)$, that is, $K(-1) = 2C_1$, $\gamma_{-1} = -3C_1$, $c(\gamma_{-1}) = C_1$, $d_w = 2 + 2C_1$. Figure 1 shows a corresponding typical realization with continuous (in scale) multifractals [*Schertzer and Lovejoy*, 1987; *Wilson*, 1991; *Pecknold et al.*, 1993]. Consider a particle injected into the center of this field (periodic boundary conditions). After 121,810 steps

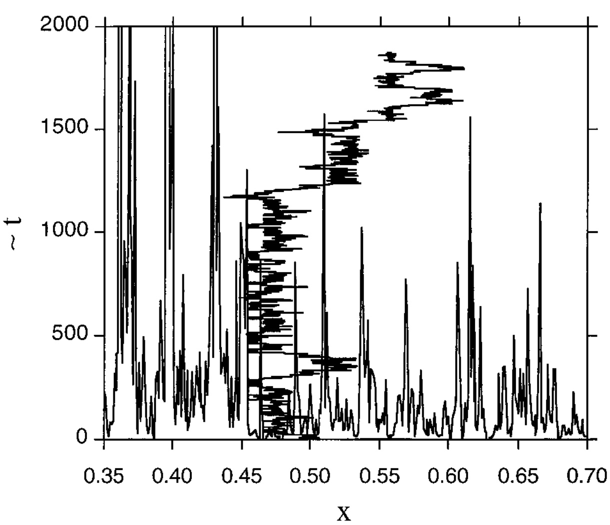

Figure 2. A random walk performed on a one-dimensional multifractal field with $\alpha = 2$, $C_1 = 0.2$, and $\lambda = 1024$. A superposition of the trail of the walk (121,080 steps taken) upon the region of the field explored by the walker (boxed region in Figure 1) is pictured here. The walker is delayed between large values of the field, hence a slow-down of the diffusion process.

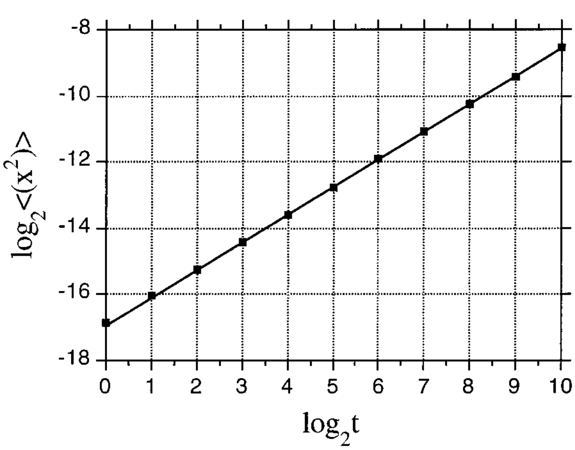

Figure 3. The mean square distance with time for $C_1 = 0.2$ and $\lambda = 1024$ averaged over 10^4 realizations and 10 particles per realization. Here $S(2) = 0.837 \pm 0.002$; theory gives 0.833.

the walker is still contained within the inset (Figure 2). It is clear from this figure that the diffusion is slowed because of the delaying of the walker between large values of the field (low-diffusivity regions).

The scaling exponent of the second-order moment $S(2)$ for diffusion in this medium is determined from Figure 3. Statistics for 10,000 realizations of the field upon each of which 10 particles were made to walk for a time of 2^{10} units yield $S(2) \cong 0.837 \pm 0.002$ (compare the theoretical value $(1 + C_1)^{-1} = 5/6 = 0.833$). The scaling exponent and its error have been determined from a least squares fit to the straight line segment. For short walks the scaling will be poor since our results were derived assuming long enough times for many sites to have been visited. For long walks there will also be a break because of the finite size of the system; however, walk lengths are limited by computer time; here they rarely exceeded a few percent of the whole system.

The "gap" (mono)scaling of the moments is illustrated by Figure 4. The straight line, whose slope is the gap exponent, confirms the monofractal nature of the walk. Here $H_w = 0.412 \pm 0.002$ and this value differs only by approximately 1% from the theoretical value (for $C_1 = 0.2$) of $H_w = 5/12 = 0.416$. Finally, Figure 5 shows good agreement between the numerical and theoretical dependence of $S(2)$ on C_1, although because of the rapid increase of trapping times with increasing C_1, the numerics become less reliable at large C_1.

It is of interest to note that although some information is available for higher spatial dimensions the situation is not clear. Qualitatively, we should expect the "barriers" to be less significant since in two or higher spatial dimensions, it is always possible to move around them; we expect the diffusion exponents to be closer to normal values $d_w = 2$, $S(2) = 1$. Indeed, *Marguerite et al.* [1998] have numerically extended the methodology presented here (also with lognormal multifractals) to two dimensions obtaining values of d_w in the range 2.17 and 2.00 ($0.92 < S(2) < 1.00$) with no systematic trend with respect to C_1 (which varied from 0 to 1); they interpret the small departures from $d_w = 2$ to be within statistical noise. On the other hand *Meakin* [1987] obtained numerical results in two-dimensions indicating subdiffusion. However, his results were on random walks, not diffusion per se; he calculated the exponent relating distance and the number of steps, not distance as a function of the elapsed time as here and in work by *Marguerite et al.* [1998]. The higher-dimensional extensions of these results are therefore not obvious.

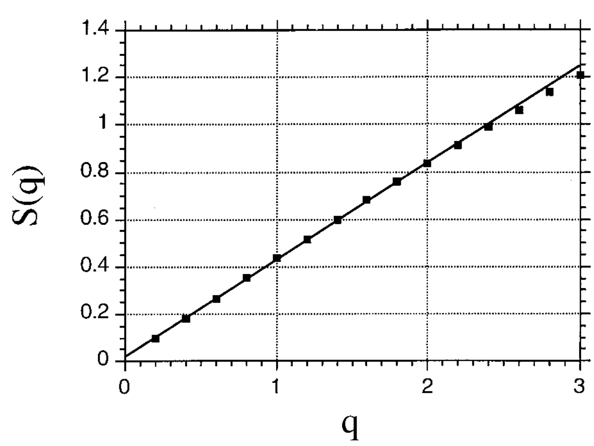

Figure 4. Scaling exponents $S(q)$ versus order of moment q. The straight line behavior indicates that the moments $\langle x^q \rangle$ can all be characterized by a single exponent H_w, where $S(q) = H_w q$. Here $H_w = 0.412 \pm 0.002$ (compare the theoretical value 0.416).

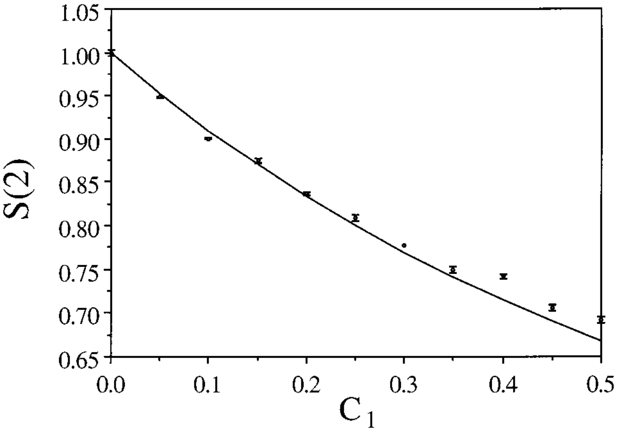

Figure 5. Dependence of the scaling exponent $S(2)$ of the second-order moment of x ($\langle x^2 \rangle \sim t^{S(2)}$) on C_1. The solid line is the theory $(1 + C_1)^{-1}$; the data points were obtained from simulations.

5. Conclusions

Diffusion on one-dimensional multifractals is the simplest paradigm for transport in scale-invariant media such as diffusion of solutes or groundwater flow through scaling porous media or of radiative transfer through scaling clouds. The corresponding statistical behavior was found to be quite rich; the fundamental difference with the standard "hierarchical" models being that singularities of all orders occur; hence we must average over different particle origins. This averaging leads to qualitatively new features associated with the statistics of the inverse (dressed) multifractal resistivity to diffusion (inverse diffusivity or inverse hydraulic conductivity). Although we obtain subdiffusive behavior with the diffusing particles being trapped in a hierarchy of barriers, in the small-scale limit of the multifractal the latter is dominated by a single order of singularity associated with a critical (fractal) set (and phase transition). Regions with higher resistivity are too rare to affect the behavior, whereas regions with lower resistivity are too weak to significantly affect the trapping.

We expect the results to be valid in one dimension when (1) $\langle \rho_\lambda \rangle$, $\langle \rho_\lambda^{-1} \rangle$ are finite and (2) when the spatial and ensemble averages of ρ_λ^{-1} are equal. Condition 1 is particularly restrictive since it is satisfied only for a single class of universal multifractals, those with Gaussian generators. Since *Liu and Molz* [1997] have found empirical evidence that hydraulic conductivities are in a different universality class, we intend to pursue study of the other universal multifractals (with other Levy generators) elsewhere. The situation for radiative transfer in clouds is less clear since although *Lovejoy and Schertzer* [1995] found statistics very close to lognormal multifractals for cloud liquid water densities, it is possible that this result could be explained by inadequate sensor response, and, although this is only suggestive, *Tessier et al.* [1993] found that cloud radiances were not lognormal. In any case, it is unlikely that either hydraulic conductivities, diffusivities or cloud densities can be modeled by the simplest case of scale-by-scale "conservative" multifractals considered here. Indeed, an additional fractional integration (power law filter) is certainly necessary for modelling cloud densities (and hence diffusivities) in clouds but probably also for hydraulic conductivities (*Liu and Molz* [1997] find the nonconservation/fractional integration parameter $H \approx 0.4$ for their logarithm; I. Tchiguirinskaia and F. Molz (manuscript in preparation, 1998) find $H \approx 0.25$ for conductivities, which is very close to that of cloud densities: $H \approx 0.3$). Since the effect of the fractional integration will be to smooth out the corresponding field, it could potentially have a drastic effect on the diffusion process. A final application suggested by the ubiquity of scaling of various rock properties [e.g., *Leary*, 1997] is the diffusion of heat in rock where we seek the distribution of temperature; in this case D is the thermal diffusivity.

Other extensions of this work include study of two- and three-dimensional systems. In the time-varying diffusion equation this includes clarification of the distinction between the random walk and the diffusion process underlined by the findings of *Marguerite et al*. [1998]: the variance of the distance traveled by a particle can no longer detect an anomalous diffusion, and a multifractal analysis is indispensible. In two or higher dimensions, several problems which were essentially trivial in one dimension become interesting; this includes the steady state diffusion problem on a multifractal or the problem of solute transport with advection given by Darcy's law with multifractal conductivities/resistivities. Finally, recent results by B. Watson et al. (manuscript in preparation, 1998) indicate that in multifractals, the connection between diffusive and radiative/kinetic transport is nontrivial, even in very thick clouds. This is because even in multifractal clouds with high average density ρ, large low-density regions can exist where photons can travel long distances with a low probability of scattering. The nonclassical nature of the multifractal statistics underscores the need for reevaluating the potentially highly nonclassical links/relationships between various types of transport processes in multifractal media.

Appendix A: Spectral Properties of *D*

In order to put the multifractal long-range correlations in a more classical spectral framework, we may consider the energy spectrum $E(k)$ of $D(x)$ (k is a wavenumber), which because of the scaling will be of the form $k^{-\beta}$. For stochastic diffusive conductances/permeabilities, *Marle et al*. [1967] and *Matheron and de Marsily* [1980] derive results which are valid only when the integral of $k^{-2}E(k)$ converges at the origin, that is, when $\beta < -1$. By evaluating β for the multifractal processes considered here, we show that they are outside the scope of the above cited stochastic methods.

For multiplicative cascade processes, the standard result [*Monin and Yaglom*, 1975] is $\beta = 1 - K_D(2)$ where $K_D(q)$ is the moment-scaling exponent of $D(x)$; hence the condition $k^{-2}E(k)$ is equivalent to $K_D(2) > 2$. To obtain $K_D(2)$ we appeal to the general result [*Lavallée et al*., 1992]:

$$\langle(\rho_\Lambda^\eta)_\lambda^q\rangle = \lambda^{K(q,\eta)} \tag{12}$$

with

$$K(q,\ \eta) = K(q\eta) - qK(\eta) \tag{13}$$

The abbreviated notation in (12) indicates that the resistance at the finest resolution Λ is raised to the η power and spatially averaged over a scale λ, and then the q power is ensemble averaged. First, we consider the upper bound on β for a conservative cascade (i.e., the direct result of multiplicative cascade; for nonconservative multifractals, the field must be power law filtered; see work by *Schertzer et al*. [1997] for new results on such a fractionally integrated flux model]. Equations (12) and (13) show (taking $q = 2$, $\eta = -1$) that $K_D(2) = K(2, -1) = K(-2) - 2K(-1)$. Since $K(q)$ is convex, this implies that $K_D(2) > 0$ (as long as $K(-2)$, $K(-1)$ converge); hence $\beta = 1 - K_D(2) < 1$. Now consider a lower bound. First note that *Marle et al*. [1967] and *Matheron and de Marsily*'s [1980] requirement $K_D(2) > 2$ implies a very strong variability; for example, in the lognormal simulations discussed above it implies $C_1 > 1/2$. Indeed, the required variability turns out to be so strong that the spectrum does not even converge. To see this, we use the fact that for dressed/integrated multifractal processes, only moments q satisfying the following inequality converge:

$$K_D(q) < d(q-1) \tag{14}$$

where d is the dimension of the space over which the dressing occurs [*Schertzer and Lovejoy*, 1987]. Taking $d = 1$, $q = 2$ in (14), we find that for convergence of the variance (and hence the spectrum), we require $K_D(2) < 1$. In other words, the spectrum of the conservative multifractal process is only well defined when $1 > \beta > 0$. We conclude that even with appropriate choice of multifractal parameters, our results necessarily remain outside the scope of the stochastic approach of *Marle et al*. [1967] and *Matheron and de Marsily* [1980].

Appendix B: Conservative Conduction Processes

Since *Liu and Molz* [1997] assume that the logarithm of the conductivity may be more fundamental than the resistivity, it is worth generalizing our results for a conservative multifractal resistance field to a conservative multifractal conductance field $\sigma_\Lambda = 1/\rho_\Lambda$. This is fairly easy to with the help of (12) and (13). Specifically, take σ_Λ as the direct result of a multiplicative process with $\langle\sigma\rangle$ = constant/independent of scale (rather than $\langle\rho\rangle$ = constant, as above). From (8) and (9) we obtain

$$\langle D_\lambda\rangle \approx \langle(\sigma_\Lambda^{-1})_\lambda^{-1}\rangle \approx \lambda^{K_\sigma(-1)} \tag{15}$$

The far right equality comes from (13); $K_\sigma(q)$ is the moment-scaling exponent for the conductivity and $K_\sigma(-1, -1) = K_\sigma(1) + K_\sigma(-1) = K_\sigma(-1)$ (since $K_\sigma(1) = 0$ for a conservative process). Equations (10) and (15) show that the formula for d_w is the same but with $K_\sigma(-1)$ in place of $K(-1)$.

Appendix C: Numerical Investigation of the Phase Transition at $\gamma_{-1} = K'(-1)$

In section 4 we found that the (anomalous) diffusion depended on $K(-1)$, that is, the -1 moment of ρ. Since in multifractals there is a one-to-one relationship between moments and field values (singularities), we noted that there will be a critical order of singularity controlling the diffusion process: $\gamma_{-1} = K'(-1)$; for the lognormal multifractal, $\gamma_{-1} = -3C_1$. To discover the effect of the various orders of singularity on the diffusion (in the limit of an infinitely large range of scales, $\lambda \to \infty$), the systematic elimination of the individual orders of singularity is performed in either of two ways. The thresholds may be imposed in such a way that the orders of singularity become bounded from above, that is, singularities of the field are removed one by one, starting with the largest and progressing to lower ones. Alternatively, the orders of singularity may be bounded from below; this is the case when first imposing a threshold at the lowest order of singularity and then progressing to higher ones. Note that this thresholding is performed on each realization of the field and that each time a threshold is imposed, the random walk process is repeated so that the statistics at these new thresholds may be monitored.

The actual thresholding procedure is the following. Consider the case where on each realization of the field the thresholding is such that the singularities are bounded from above. Once the first threshold has been determined and imposed, any region of the field with a value that exceeds it is assigned the threshold value. Clearly, for different threshold values of a particular field there will be different rates of diffusion, and therefore the asymptotic regime, from which the scaling exponents are determined, will be attained at different times. For instance, eliminating large singularities from the field will cause the diffusion to be more rapid. Therefore each time a threshold is imposed and before the random walk process is repeated the field is normalized, for the sake of numerical simplicity, such that $\bar{\rho}_\lambda = 1$. Normalizing the field (multiplying it by a constant) does not affect the diffusion exponents; it merely rescales the timescale of the problem.

To determine how the thresholding changes the codimension function $c(\gamma)$, note that when a threshold characterized by an order of singularity γ_t is imposed (still bounding from above), all singularities which have an order greater than γ_t are eliminated and hence the probability of finding any is zero; the largest order of singularity that will then exist for the field is γ_t. The exceedence set characterized by this same γ_t, however, remains unchanged (and so does $c(\gamma_t)$ and $c(\gamma)$ for $\gamma < \gamma_t$) since all the orders of singularity that were previously larger than γ_t are set equal to γ_t. This is illustrated in Figure 6, which indicates how the bare codimension function changes for lognormal multifractals when the singularities are bounded (thresholded) from above. (Note that when $c(\gamma)$ has a minimum, as for lognormal multifractals where it is a quadratic, the definition of (1) must be extended to the left-hand side using a

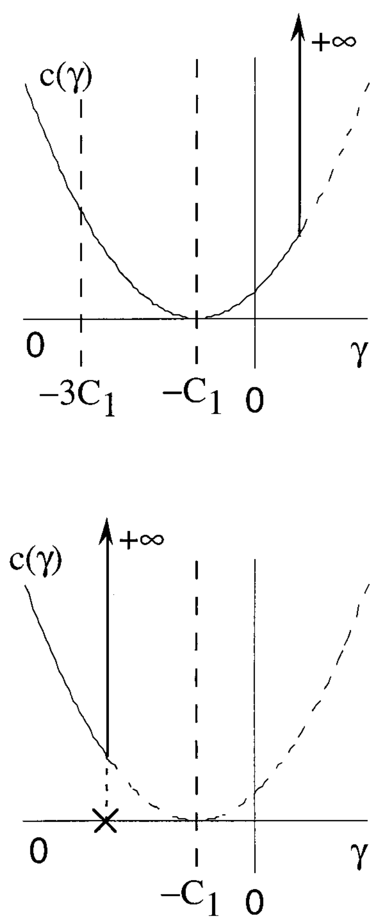

Figure 6. The effects of thresholding on $c(\gamma)$, the bare codimension function, for lognormal multifractals when bounding the orders of singularity from above.

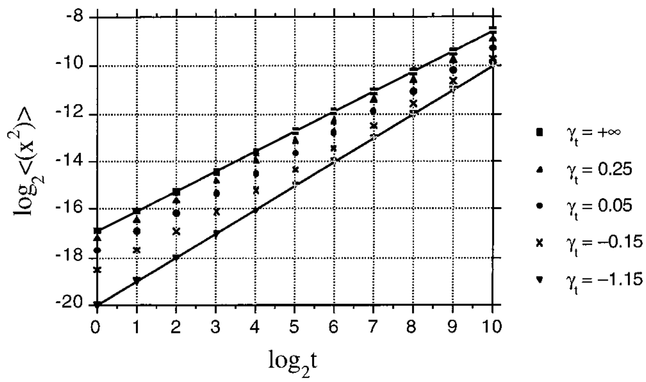

Figure 7. The effects of thresholding on the scaling of the mean square distance with time for diffusion on a one-dimensional multifractal with $C_1 = 0.2$ and $\lambda = 1024$, when bounding the orders of singularity from above. The scaling exponent $S(2)$ increases as the threshold T is lowered ($T \sim \lambda^{\gamma_t}$); when $\gamma_t = +\infty$, $S(2) = 0.837 \pm 0.002$, and when $\gamma_t = -1.15$, $S(2) = 1.0031 \pm 0.0007$.

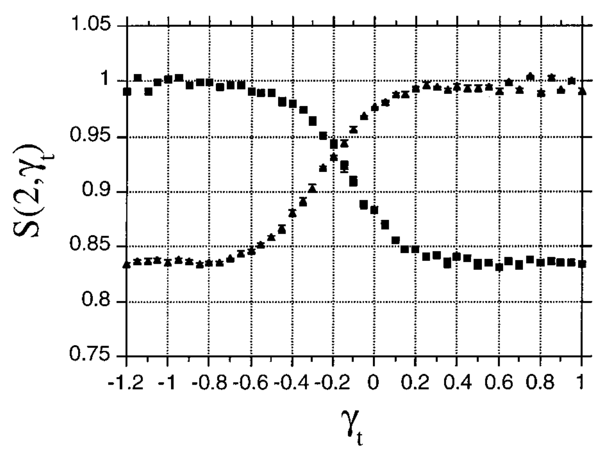

Figure 8. Scaling exponent $S(2)$ as a function of the order of singularity γ_t, which determines the threshold ($T \sim \lambda^{\gamma_t}$), for the lognormal multifractal with $C_1 = 0.2$ and $\lambda = 1024$. The squares indicate that thresholding began with the removal of the largest singularity and so on to the smaller ones; the triangles indicate that the process began with the removal of the smallest singularity and so on to the larger ones. A transition from anomalous to normal diffusion occurs about $\gamma = -0.2$; the transition region begins roughly about $\gamma = -3C_1 = -0.6$. The "smearing" of this transition is apparently a finite size effect (see Figure 9).

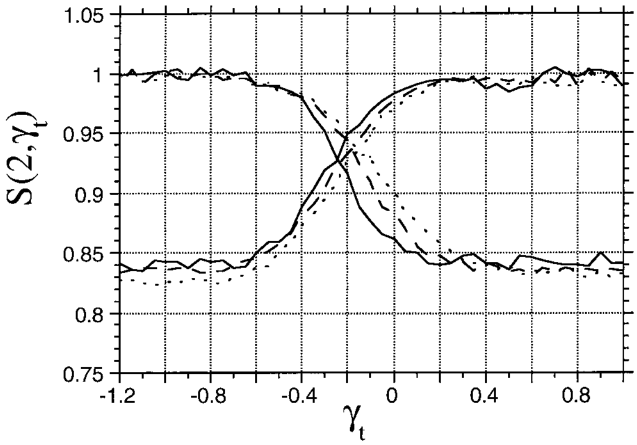

Figure 9. Same as Figure 8 but showing how the transition becomes sharper and closer to $\gamma_{-1} = -3C_1$ ($= -0.6$) as λ is increased from $\lambda = 256$ (dotted lines) to $\lambda = 1024$ (dashed lines) and $\lambda = 16384$ (solid lines). Errors are on the order of 0.005.

"$<$" rather than "$>$" in (1) for $\gamma < \gamma_0$ and $c'(\gamma_0) = 0$; thus for the lognormal case, $c(\gamma) = (\gamma + C_1)^2/(4C_1))$. The above change in $c(\gamma)$ is valid only for a large range of scales ($\lambda \to \infty$); for finite λ the thresholding (which is performed at a unique scale) breaks the scaling over a range (see work by *Larnder* [1995] for details); therefore care must be taken when looking at the statistics of the new (dressed) field.

A first glimpse of how this thresholding affects the statistics of the random walks is provided by Figure 7. This graph displays several plots of the scaling of the mean square distance with time for walks that took place on a field with $C_1 = 0.2$ and $\lambda = 1024$; each plot represents the statistics for walks on the field when a particular threshold was imposed. Here again, the singularities were bounded from above. As the threshold was lowered, the extreme singularities were eliminated; this facilitated transport throughout the field; hence the diffusion rates (the scaling exponents) increased.

In order to study more clearly the behavior of the random walkers, the scaling exponent of the second moment $S(2)$ was plotted as a function of the threshold singularity γ_t. The entire procedure was executed using both methods of thresholding for several different values of C_1. For a given C_1 the results for $S(2, \gamma_t)$ that were obtained when the singularities of the field were bounded from above were superimposed with those obtained when the singularities were bounded from below. Figure 8 displays the two plots of $S(2, \gamma_t)$ versus the order of singularity γ_t, which characterizes the threshold, for random walks on a multifractal field with $C_1 = 0.2$ and $\lambda = 1024$. These plots indicate a transition from anomalous to normal diffusion; the transition region begins roughly about the anticipated order of singularity, $\gamma_{-1} = -3C_1 = -0.6$, and the transition itself is centered roughly about $\gamma = -0.2$. Toward the end of the thresholding process in both plots there occurs a slight fluctuation about $S(2) = 1$, the value of the scaling exponent for normal diffusion. These statistical fluctuations could be reduced by allowing longer walks to take place; longer walks would provide more points for the scaling regime, from which the scaling exponent is determined. The transition that is observed in Figure 8 appears smeared (it is not sharp). Figure 9 demonstrates that the smearing is a finite size effect; for systems with smaller λ the transition region is broader, and for systems which have larger λ the transition is clearly steeper. Furthermore, as λ is increased, the point at which the transition is centered moves steadily (although the displacement is slight) toward smaller order singularities yet the transition region always seems to begin at $\gamma = -0.6$. Therefore, although the convergence is slow, it is plausible that in the limit $\lambda \to \infty$ there is a "dynamical phase transition" about $\gamma_{-1} = -3C_1$.

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S. Lovejoy and P. Silas, Department of Physics, McGill University, 3600 University St., Montréal, Québec H3A 2T8, Canada. (e-mail: lovejoy@physics.mcgill.ca)

D. Schertzer, LMM (CNRS) Boite 162, Université P. & M. Curie, 4 Pl. Jussieu, Paris 75252, Cedex 05, France.

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