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Multiple Logarithmic Oscillations for Statistical Fractals on Ultrametric Spaces with Application to Recycle Flows in Hierarchical Porous Media

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Abstract

The interference of logarithmic oscillations characteristic to statistical fractal and ultrametric structures is analyzed. We introduce an ultrametric model consisting in a hierarchy of branches for which the probability distribution $\xi(n)$ of the total number, n, of branches has a long tail of the inverse power law type $\xi(n) \sim A(\ln n)n^{-(1+H)}$ as $n \to \infty$ where H is a fractal exponent characteristic for the ultrametric structure and $A(\ln n)$ is a periodic function of ln n. A comparison is made with a statistical fractal derived by means of the Shlesinger-Hughes stochastic renormalization approach. We consider a positive random variable, X, selected from a narrow unimodal probability density with finite moments. A process of stochastic amplification of the random variable is introduced which leads to a probability density of the amplified variable with a long tail: $\tilde{P}(X) dX \sim dX$ $X^{-(1+\mathscr{H})}B[\ln(X/X_m)]$ as $X \to \infty$ where \mathscr{H} is another fractal exponent, X_m is a cutoff value of the random variable X and $B[\ln (X/X_m)]$ is a periodic function of $\ln (X/X_m)$. Finally, the interaction between these two types of logarithmic oscillations is analyzed by assuming that the stochastic amplification of the random variable X takes place on an ultrametric structure. The final probability density of X, $P^*(X) dX$, displays a phenomenon of amplitude modulation $P^*(X) dX \sim d[\ln (X/X_m)] [\ln (X/X_m)]^{-(1+H)} \Xi$ as $X \to \infty$ where $\Xi = \Xi^{(1)} + \Xi^{(2)}$ is made up of two additive contributions: a periodic function $\Xi^{(1)}[\ln (X/X_m)]$ of $\ln (X/X_m)$ and a superposition $\Xi^{(2)}$ {ln (X/X_m) , ln [ln (X/X_m)] of periodic functions of ln (X/X_m) modulated by much slower periodic functions in ln $[\ln (X/X_m)]$. The model is applied for the analysis of recycle flows in porous media. We assume that a hierarchical structure of pores exists which corresponds to an ultrametric space and that the recycle flow leads to a stochastic amplification of the residence time of fluid elements in the system. We show that the probability density of the residence time has an asymptotic behavior similar to that of $P^*(X)$ dX and investigate the possibilities of measurement of multiple logarithmic oscillations by means of tracer experiments. A physical interpretation of multiple logarithmic oscillations is given in the case of flow systems: they are generated by two delayed feedback processes occurring in two different logarithmic time scales, $\ln t$ and $\ln \ln t$. The fast delayed feedback process in ln t is given by the recycle flows corresponding to a given level of the porous structure whereas the slow delayed feedback process in ln ln t is due to the exchange of fluid among the different levels of the hierarchical structure of pores.

1. Introduction

It is now well established that the probability distributions corresponding to the statistical fractal or ultrametric structures may have long tails of the inverse power law type modulated by oscillatory functions depending on the logarithm of the random variable (Novikov [1], Schrekenberg [2], Giacometti, Maritan and Stella [3] and references therein, West [4, 5] and references therein, Shlesinger [6] and references therein). Such logarithmic oscillations have been experimentally identified in many biological as well as physical systems (Anselmet *et al.* [7], Smith, Fournier and Spiegel [8], West, Bhargava and Goldberger [9], Nelson, West and Goldberger [10], Shlesinger and West [11]).

This note addresses a problem which has not been investigated yet, the analysis of interactions between two such logarithmic oscillatory processes. In order to build a model which incorporates two different logarithmic oscillations we shall apply the stochastic renormalization procedure of Shlesinger and Hughes [12] to an ultrametric model introduced by Vlad [13, 14]. As a simple physical example of the interaction between two logarithmic oscillatory processes we consider the recycle flow in porous media with a hierarchical structure described in terms of the probability density of residence times (Nauman and Buffham [15] and references therein). This example from hydrodynamics has two main advantages: it is amenable to analytical treatment and on the other hand it may be studied by means of tracer experiments.

2. Ultrametric topology

In this section an ultrametric structure is constructed by using a method suggested by Vlad [13, 14]. We assume that the ultrametric structure is generated by a branching process with a random number of branches. The process of branches generation is similar to a hierarchical clustering. We suppose that the probability β of a branch generation is constant. Thus the probability that a branch from the (q-1)th level is connected to *n* branches from the *q*th level is $\beta^{n-1}(1-\beta)$. If there are *n'* branches at the (q-1)th level the probability $\phi_{n'}(n)$ that there are *n* branches at the *q*th level is equal to

$$\phi_{n'}(n) = \sum_{n_1} \cdots \sum_{n_{n'}} \delta_{n(n_1 + \cdots + n_{n'})} \prod_{l=1}^{n'} [\beta^{n_l - 1}(1 - \beta)]$$

= $\beta^{n-n'}(1 - \beta)^{n'}(n - 1)!/[(n' - 1)!(n - n')!], \quad n \ge n'.$

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The probability, $\xi_o(n)$, that there are *n* branches at the *q*th tively level can be computed step by step

$$\xi_q(n) = \sum_{n \ge n'} \phi_{n'}(n) \xi_{q-1}(n'), \quad \xi_0(n) = \delta_{n1}.$$
 (2)

By solving these equations recursively we get

$$\xi_q(n) = (1 - \beta)^q [1 - (1 - \beta)^q]^{n-1}.$$
(3)

Now we introduce the probability, α , that the growth of a level of the hierarchy occurs. The probability, χ_a , that the growth of the hierarchy stops after q steps is equal to

$$\chi_q = \alpha^q (1 - \alpha). \tag{4}$$

The probability $\tilde{\xi}(n)$ that the total number of branches from a hierarchy is n may be computed as an average of $\xi_n(n)$ over the possible number of levels

$$\tilde{\xi}(n) = \sum_{q=0}^{\infty} \chi_q \, \xi_q(n). \tag{5}$$

By inserting eqs (3) and (4) into eq. (5) we obtain

$$\tilde{\xi}(n) = \sum_{q=0}^{\infty} \alpha^{q} (1-\alpha)(1-\beta)^{q} [1-(1-\beta)^{q}]^{n-1}.$$
(6)

Equation (6) is typical for an ultrametric model. Each contribution of a level to $\tilde{\xi}(n)$ is an exponential probability distribution in n [eq. (3)]; the corresponding average values

$$\langle n \rangle_q = \sum_{n=1}^{\infty} n \xi_q(n) = (1-\beta)^{-q}, \tag{7}$$

increase exponentially with the level index, q. On the contrary, the corresponding weight function $\alpha^q(1-\alpha)$ decreases exponentially as q increases. The interplay between these two factors may lead to the absence of a characteristic scale of the system generating a statistical fractal. Indeed, by evaluating the asymptotic behavior of eq. (6) by means of the Poisson summation formula [16] we get

$$\tilde{\xi}(n) \sim n^{-(1+H)} A(\ln n) \quad \text{as } n \to \infty, \tag{8}$$

where H is a fractal exponent given by

$$H = \ln \alpha / \ln (1 - \beta), \tag{9}$$

 $A(\ln n)$ is a periodic function of $\ln n$ with period $-\ln(1-\beta)$:

$$A(\ln n) = \frac{1-\alpha}{-\ln (1-\beta)} \left\{ \Gamma(1+H) + 2\sum_{m=1}^{\infty} \left[F^+ \left(1+H, \frac{2\pi m}{-\ln (1-\beta)} \right) \right] \times \cos \left(\frac{2\pi m (\ln n)}{-\ln (1-\beta)} + F^- \left(1+H, \frac{2\pi m}{-\ln (1-\beta)} \right) \right] \times \sin \left(\frac{2\pi m (\ln n)}{-\ln (1-\beta)} \right) \right\},$$
(10)

 $\Gamma(x) = \int_0^\infty y^{x-1} \exp(-y) \, dy, \quad x > 0$ is the complete gamma function and $F^{\pm}(a, b)$ are the real and imaginary parts of the gamma function of complex argument, respec-

$$F^{\pm}(a, b) = \begin{cases} \operatorname{Re} \\ \operatorname{Im} \end{cases} \Gamma(z = a + ib) \\ = \int_{0}^{\infty} e^{-y} y^{a-1} \begin{cases} \cos \\ \sin \end{cases} (b \ln y) \, dy.$$
(11)

3. Statistical fractals

We consider a positive random variable, X, selected from a narrow, unimodal probability density, $P_0(X) dX$, with finite positive moments. A probability distribution, $\tilde{P}(X) dX$, with a long tail may be generated by applying the Shlesinger-Hughes renormalization procedure (Shlesinger and Hughes [12]). We assume that the random variable X is subject to a multi-step amplification process characterized by an amplification factor b > 1. We have

$$\widetilde{P}(X) = \sum_{n=0}^{\infty} \varepsilon_n \int \delta(X'b^n - X) P_0(X') \, \mathrm{d}X', \qquad (12)$$

where ε_n is the probability that a cascade of amplification processes is made up of n amplification events; it is given by:

$$\varepsilon_n = \lambda^n (1 - \lambda), \tag{13}$$

where λ is the probability that an amplification event occurs. By inserting eq. (13) into eq. (12), computing the integral over X' and evaluating the resulting sum by means of the Poisson formula we get the following expression for the asymptotic behavior of $\tilde{P}(X)$

$$\tilde{P}(X) \cong X^{-(1+\mathscr{H})} B(\ln X) \quad \text{as } X \to \infty, \tag{14}$$

where \mathcal{H} is a fractal exponent similar to H:

$$\mathscr{H} = \ln (1 - \lambda) / \ln b, \tag{15}$$

and $B(\ln X)$ is a periodic function of $\ln X$ with period $\ln b$:

$$B(\ln X) = [(1 - \lambda)/\ln b] \left(\langle X_0^{\mathscr{H}} \rangle + 2 \sum_{m=1}^{\infty} \{ \langle X_0^{\mathscr{H}} \cos [(2\pi m \ln X_0)/\ln b] \rangle \times \cos [(2\pi m \ln X)/\ln b] + \langle X_0^{\mathscr{H}} \sin [(2\pi m \ln X_0)/\ln b] \rangle \times \sin [(2\pi m \ln X)/\ln b] \} \right).$$
(16)

Here the average over X_0 is computed in terms of $P_0(X_0)$ dX_0 :

$$\langle \cdots \rangle = \int P_0(X_0) \cdots dX_0.$$
 (17)

If there is a cutoff value, X_m , of X for which

$$P_0(X > X_m) \approx 0, \tag{18}$$

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then eq. (16) becomes

$$B[\ln (X/X_{m})] = [(1 - \lambda)/\ln b] \left(\langle (X_{0})^{\mathscr{H}} \rangle + 2 \sum_{m=1}^{\infty} (\langle (X_{0})^{\mathscr{H}} \cos \{ [2\pi m \ln (X_{0}/X_{m})]/\ln b \} \rangle \times \cos \{ [2\pi m \ln (X/X_{m})]/\ln b \} + \langle (X_{0})^{\mathscr{H}} \sin \{ [2\pi m \ln (X_{0}/X_{m})]/\ln b \} \rangle \times \sin \{ [2\pi m \ln (X/X_{m})]/\ln b \}) \right).$$
(19)

4. Statistical fractals on ultrametric structures

Now we can combine the two approaches. We assume that to each new branch of the ultrametric structure there corresponds an amplification event; in other words if the ultrametric structure has only one branch then there is no amplification; for two branches, there is an amplification event; in general, for *n* branches there are n - 1 amplification events. It follows that the probability density of *X*, $P^*(X) dX$, attached to the whole hierarchy, is given by

$$P^{*}(X) = \sum_{n=1}^{\infty} \tilde{\xi}(n) \int \delta(X - X'b^{n-1}) P_{0}(X') \, \mathrm{d}X'.$$
 (20)

By using eqs (3)–(6) we can express $P^*(X)$ as a sum of contributions corresponding to the different levels of the ultrametric structure

$$P^{*}(X) = \sum_{n=1}^{\infty} \sum_{q=0}^{\infty} (1-\alpha)\alpha^{q}(1-\beta)^{q} \times [1-(1-\beta)^{q}]^{n-1}b^{1-n}P_{0}(Xb^{1-n})$$
$$= \sum_{q=0}^{\infty} (1-\alpha)\alpha^{q}\tilde{P}_{q}(X), \qquad (21)$$

where

$$\tilde{P}_{q}(X) = \sum_{n=1}^{\infty} (1-\beta)^{q} [1-(1-\beta)^{q}]^{n-1} b^{1-n} P_{0}(Xb^{1-n}), \quad (22)$$

is the probability density of X corresponding to the *q*th level of the ultrametric structure. By applying the Poisson method we get the following expression for the asymptotic behavior of $\tilde{P}_{a}(X)$

$$\tilde{P}_{q}(X) \cong X^{-(1+\mathscr{H}_{q})}B_{q}(\ln X) \quad \text{as } X \to \infty.$$
(23)

To each level corresponds a fractal exponent

$$\mathscr{H}_{q} = -\ln\left[(1-\beta)^{q}\right]/\ln b, \qquad (24)$$

and a periodic function of $\ln X$ with period $\ln b$:

$$B_{q}(\ln X) = [(1 - \beta)^{q}/\ln b] \Big(\langle (X_{0})^{\mathscr{H}_{q}} \rangle \\ + 2 \sum_{k=1}^{\infty} \{ \langle (X_{0})^{\mathscr{H}_{q}} \cos [(2\pi k \ln X_{0})/\ln b] \rangle \\ \times \cos [(2\pi k \ln X)/\ln b] \\ + \langle (X_{0})^{\mathscr{H}_{q}} \sin [(2\pi k \ln X_{0})/\ln b] \rangle \\ \times \sin [(2\pi k \ln X)/\ln b] \} \Big)$$
(25)

For $\beta < 1$ even a moderate value of the level index q (say 4 or 5) leads to $(1 - \beta)^q \ll 1$ and, thus, a very good approximation for \mathcal{H}_q is

$$\mathscr{H}_{q} \cong (1-\beta)^{q}/\ln b. \tag{25}$$

By combining eqs (21), (23), (25) and (26) and applying again the Poisson summation formula we get the following asymptotic expression for $P^*(X)$ as $X \to \infty$:

$$P^{*}(X) \cong \frac{1-\alpha}{X (\ln b)[-\ln (1-\beta)]} \times \left\{ \left\langle \left(\frac{\ln b}{\ln (X/X_{0})}\right)^{H+1} \right\rangle \Gamma(1+H) + 2\Gamma(1+H) \sum_{k=1}^{\infty} \left[\left\langle \left(\frac{\ln b}{\ln (X/X_{0})}\right)^{H+1} \right\rangle \times \cos \left(\frac{2\pi k \ln (X/X_{0})}{\ln b}\right) \right\rangle \right] + 4 \sum_{k=1}^{\infty} \sum_{m=1}^{\infty} \left\{ \left\langle \left(\frac{\ln b}{\ln (X/X_{0})}\right)^{H+1} \cos \left(\frac{2\pi k \ln (X/X_{0})}{\ln b}\right) \right\rangle \times \left[F^{+} \left(H+1, \frac{2\pi m}{-\ln (1-\beta)}\right) \right] \times \cos \left(\frac{2\pi m \ln [\ln (X/X_{0})/\ln b]}{-\ln (1-\beta)}\right) + F^{-} \left(H+1, \frac{2\pi m}{-\ln (1-\beta)}\right) \right\} \times \sin \left(\frac{2\pi m \ln [\ln (X/X_{0})/\ln b]}{-\ln (1-\beta)}\right) \right\},$$
as $X \to \infty$, (27)

where the average over X_0 is computed by using the probability distribution $P_0(X_0)$, H is the fractal exponent attached to the ultrametric structure [eq. (9)] and the other variables have the same significance as before.

It is difficult to analyse eq. (27) in the general case; however, if the approximation (18) is valid we get a simpler expression for $P^*(X)$ as $X \to \infty$:

$$P^*(X) dX = \Xi[\ln (X/X_m)]^{-(H+1)} d[\ln (X/X_m)]$$

as $X \to \infty$, (28)
where

$$\Xi = \Xi^{(1)} + \Xi^{(2)},\tag{29}$$

is a slowly varying function of X made up of two contributions

$$\Xi^{(1)}[\ln (X/X_{\rm m})] = \frac{(1-\alpha)(\ln b)^{\rm H}}{-\ln (1-\beta)}$$

$$\times \Gamma(1+H) \left\{ 1 + 2\sum_{k=1}^{\infty} \left[\cos \left(\frac{2\pi k \ln (X/X_{\rm m})}{\ln b} \right) \right]$$

$$\times \left\langle \cos \left(\frac{2\pi k \ln (X_0/X_{\rm m})}{\ln b} \right) \right\rangle$$

$$+ \sin \left(\frac{2\pi k \ln (X/X_{\rm m})}{\ln b} \right)$$

$$\times \left\langle \sin \left(\frac{2\pi k \ln (X_0/X_{\rm m})}{\ln b} \right) \right\rangle$$
(30)

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$$\begin{split} \Xi^{(2)}[\ln (X/X_{m}), \ln (\ln (X/X_{m})] \\ &= \frac{4(1-\alpha)(\ln b)^{H}}{-\ln (1-\beta)} \sum_{k=1}^{\infty} \left\{ \left[\cos \left(\frac{2\pi k \ln (X/X_{m})}{\ln b} \right) \right. \right. \\ &\times \left\langle \cos \left(\frac{2\pi k \ln (X_{0}/X_{m})}{\ln b} \right) \right\rangle \\ &+ \sin \left(\frac{2\pi k \ln (X/X_{m})}{\ln b} \right) \left\langle \sin \left(\frac{2\pi k \ln (X_{0}/X_{m})}{\ln b} \right) \right\rangle \right] \right\} \\ &\times \left\{ \sum_{m=1}^{\infty} \left[F^{+} \left(H + 1, \frac{2\pi m}{-\ln (1-\beta)} \right) \\ &\times \cos \left(\frac{2\pi m \ln [\ln (X/X_{m})/\ln b]}{-\ln (1-\beta)} \right) \\ &+ F^{-} \left(H + 1, \frac{2\pi m}{-\ln (1-\beta)} \right) \\ &\times \sin \left(\frac{2\pi m \ln [\ln (X/X_{m})/\ln b]}{-\ln (1-\beta)} \right) \right] \right\}, \\ &X = X_{m}, X \to \infty. \end{split}$$

 $\Xi^{(1)}$ is a periodic function of $\ln (X/X_m)$ with period $\ln b$; it has the main contribution to Ξ . $\Xi^{(2)}$ is the sum of a set of periodic functions of $\ln (X/X_m)$ with period $\ln b$ modulated by much slower periodic functions in $\ln[\ln (X/X_m)]$ with period $-\ln (1 - \beta)$.

5. Recycle flows in open systems

Residence time distribution measurements have been used in engineering for 50 years (Nauman and Buffham [15] and references therein). The mathematical formalism used for the interpretation of experimental data is somewhat similar to some models used in statistical physics, e.g. with the random walk theory. Although several papers outlining the analogies between these two fields have already been published (Rappaport and Dayan [17], Schweich [18], Van den Broeck [19], Vlad [20-22]), the formalism of residence time distributions is almost unkown by physicists.

Our purpose is to describe the residence time distributions for recycle flow in porous media by using the formalism developed in the preceding section. The first step of our analysis is the study of recycle flows in homogeneous systems without pores. We shall show that the recycling of a fluid in a given region of space may result in stochastic amplification of residence time provided that certain conditions are fulfilled.

We use a simplified version of a model developed by Vlad [22]. We consider an open continuous flow system and assume that the fluid is incompressible and that the outlet rate is the same as the inlet rate. The flow process is made up of many cycles $m = 0, 1, 2, \ldots$. For each cycle, *m*, we can introduce a probability density of the residence time of a fluid element in the system

$$\Phi_m(\tau) d\tau = \text{independent of time}, \int_0^\infty \Phi_m(\tau) d\tau = 1;$$
 (32)

 $\Phi_m(\tau) d\tau$ is the probability that a fluid molecule entering the cycle at time t will leave it at a time between $t + \tau$ and $t + \tau + d\tau$, i.e. that the residence time corresponding to a given cycle is between τ and $\tau + d\tau$. We assume that all

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moments of the residence time corresponding to a cycle exist and are finite. In particular the average residence times

$$\theta_m = \int_0^\infty \tau \Phi_m(\tau) \, \mathrm{d}\tau, \tag{33}$$

may be expressed as [15, 22]:

$$\theta_m = V_m / Q_m, \tag{34}$$

where V_m is the volume available for the *m*th flow cycle and Q_m is the corresponding flow rate. We assume that all flow cycles are similar to each other so that Φ_m obey the scaling law [15, 22]

$$\Phi_m(\tau) = (\theta_m)^{-1} \eta(\tau/\theta_m), \tag{35}$$

where the probability density $\eta(x) dx$ of the dimensionless residence time, $x = \tau/\theta_m$, is the same for all cycles. The scaling law (35) is commonly used in the literature [15, 22, 23].

We consider that from cycle to cycle a bigger and bigger volume is involved in the flow process with a smaller and smaller flow rate. For two successive pairs of values V_{m-1} , Q_{m-1} and V_m , Q_m we have the relationships [22]

$$V_m = \mu_m V_{m-1}, \quad Q_m = a_m Q_{m-1}$$
 (36)

where $\mu_m > 1$, m = 1, 2, ... are expansion coefficients of the volumes involved in different flow cycles and a_m is the fraction of the flow rate, Q_{m-1} corresponding to the (m-1)th cycle, involved in the *m*th cycle. Following Vlad [22] we assume that the pairs $(a_1, \mu_1), (a_2, \mu_2), ...$ are independent random variables selected from the same probability law

$$\sigma(a, \mu) \,\mathrm{d}a \,\mathrm{d}\mu, \, \int_{1}^{\infty} \,\int_{0}^{1} \sigma(a, \mu) \,\mathrm{d}a \,\mathrm{d}\mu = 1.$$
 (37)

By combining eqs (34)-(36) we get the following expressions for the flow rates, Q_m , and average residence times, θ_m , corresponding to the different cycles

$$Q_0 = (1 - a_1)Q, Q_m = (1 - a_{m+1})a_m \cdots a_1Q,$$
 (38)

and

$$\theta_m = (\mu_m \cdots \mu_1 / a_m \cdots a_1) \theta_0, \qquad (39)$$

where

$$Q = \sum_{m=0}^{\infty} Q_m, \qquad (40)$$

is the total flow rate and θ_0 is the average residence time corresponding to the 0th cycle.

The above hypotheses concerning the flow mechanism are identical with the ones used by Vlad [22]. Now we introduce a supplementary assumption which is different from the one used in Ref. [22]. We assume that the recycle flow takes place within the system and that a fluid element leaving the system can never return; in the engineering language we deal with internal backmixing. In contrast, Ref. [22] discusses the case of external backmixing, that is, it is assumed that for each cycle a certain amount of fluid leaving the system can return into it.

For internal backmixing the overall probability distribution $\psi(\tau) d\tau$ of the residence time in the system can be evaluated as an average over the number of cycles and over the values of the random parameters $(a_1, \mu_1), (a_2, \mu_2), \ldots$:

$$\psi(\tau) = \sum_{m=0}^{\infty} \int_{0}^{1} \int_{1}^{\infty} \cdots \int_{0}^{1} \int_{1}^{\infty} (Q_{m}/Q) \Phi_{m}(\tau) \times \prod_{m'=1}^{m+1} [\sigma(a_{m'}, \mu_{m'}) \, \mathrm{d}a_{m'} \mathrm{d}\mu_{m'}].$$
(41)

After some arrangements eq. (41) can be written as

$$\psi(\tau) = \int_{0}^{1} \int_{1}^{\infty} (1-a) \Phi_{0}(\tau) f(a, b) \, da \, db$$

+ $\sum_{m=1}^{\infty} \int_{0}^{1} \int_{1}^{\infty} \cdots \int_{0}^{1} \int_{1}^{\infty} (1-a_{m+1}) \frac{a_{m} \cdots a_{1}}{b_{m} \cdots b_{1}}$
× $\Phi_{0} \left(\frac{\tau}{b_{m} \cdots b_{1}}\right) f(a_{1}, b_{1}) \, da_{1} \, db_{1} \cdots$
 $\cdots f(a_{m+1}, b_{m+1}) \, da_{m+1} \, db_{m+1},$ (42)

where

$$b_m = \mu_m / a_m > 1, \quad m = 1, 2, \dots,$$
 (43)

are effective amplification factors of the total residence time corresponding to different cycles

$$f(a, b) da db = \sigma(a, \mu) da d\mu$$

= $a\sigma(a, ab) da db$, $\int_0^1 \int_1^\infty f(a, b) da db = 1$, (44)

is the probability density of the random vectors (a_1, b_1) , (a_2, b_2) , ... and $\phi_0(\tau)$ is the probability density of the residence time for the 0th cycle.

By comparing eqs (12) and (13) with eq. (42) we note that the recycle problem for homogeneous systems is equivalent to the stochastic renormalization approach presented in Section 3 provided that the random vectors $(a_1, b_1), (a_2, b_2),$... can take only one value $(a, b) = (a_0, b_0)$, that is, if the probability density f(a, b) is given by a delta function

$$f(a, b) = \delta(a - a_0)\delta(b - b_0).$$
 (45)

If the condition (45) is fulfilled then eq. (42) reduces to a relation equivalent to eqs (12) and (13)

$$\psi(\tau) = \sum_{m=0}^{\infty} (1-a)a^m \int \delta(\tau' b^m - \tau) \Phi_0(\tau') \, \mathrm{d}\tau'. \tag{46}$$

By comparing eqs (12), (13) and (46) we can derive the following "dictionary"

$$\tau \leftrightarrow X, \quad \tau' \leftrightarrow X', \tag{47}$$

$$\psi(\tau) \leftrightarrow P(X) \quad \phi_0(\tau') \to P_0(X'), \tag{48}$$

$$a_0 = a \leftrightarrow \lambda, \quad b_0 = b \leftrightarrow b.$$
 (49)

The physical interpretation of the above results is simple: each recycle event of the fluid corresponds to an event of stochastic amplification of the residence time of fluid elements in the system. For the more general process described by eq. (42) the effective amplification factors $b_m = \mu_m/a_m$ and the probabilities of occurrence of the different amplification (recycle) events a_m are random variables selected from the probability law (44). The probability that *m* amplification (recycle) events occur is given by the ratio between the flow rate corresponding to the *m*th cycle and the total flow rate,

$$\varepsilon_m = Q_m/Q = (1 - a_{m+1})a_m \cdots a_1.$$
 (50)

The process of stochastic amplification of the residence time described by eq. (42) is more general than the stochastic renormalization scheme considered in Section 3. Only if eq. (45) is valid then the parameters $(a_1, b_1), (a_2, b_2), \ldots$ are constant, the two models are equivalent and the asymptotic behavior of the probability density, $\psi(\tau) d\tau$, of the total residence time has an inverse power law asymptotic behavior modulated by logarithmic oscillations described by eqs (14)-(19) where the significance of the symbols is given by the "dictionary" (47)-(49) and by

$$\tau_{c} \leftrightarrow X_{m}; \tag{51}$$

where τ_c is the cutoff value of the residence time for which the probability density attached to the 0th cycle is practically equal to zero,

$$\Phi_0(\tau > \tau_c) \approx 0. \tag{52}$$

6. Recycle flows in hierarchical porous media

Now we can proceed to discuss the problem of backmixing in hierarchical porous media. We assume that the flow system is not homogeneous but rather filled with a porous substance in which a broad distribution of pores of different sizes exists. We consider that the pores are arranged in a hierarchical way: the large pores are connected to small pores, the small pores to smaller pores, etc. Like the branches of an ultrametric structure the pores may be grouped in levels. The pores of largest size correspond to a label q = 0 and as q increases the sizes of the pores decrease; a pore from the *q*th level is connected to one larger pore from the (q-1)th level and to a variable number of smaller pores from the (q + 1)th level. Such a hierarchy of pores which is similar to an ultrametric structure is commonly encountered in nature [24, 25]. Of course, for real systems the self-similarity typical for an ultrametric space does not act up to infinity; there is always a cutoff value for which the self-similarity no longer exists. In many cases the influence of the cutoff value on the macroscopic properties of the system is negligible. Experimental electrochemical [26] and adsorption [27, 28] measurements show that this is indeed the case.

From the hydrodynamic point of view a hierarchical structure of pores slows down the speed of flow and increases the efficiency of recycling. For a homogeneous flow system without pores and with $(\lambda, b) = (a, b) = (a_0, b_0) = constant$ the probability, ε_m , of the occurrence of m cycles is given by the Pascal law (13):

$$\varepsilon_m = (1-a)a^m. \tag{13} = (53)$$

This distribution is rather narrow and all positive moments of the number of cycles exist and are finite. A straightforward calculation gives

$$\langle m(m-1)\cdots(m-k+1)\rangle = \sum m(m-1)\cdots$$

 $(m-k+1)\varepsilon_m = k![a/(1-a)]^k.$ (54)

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On the contrary, for an ultrametric structure of pores one expects that ε_m has a long tail of the invese power law type and all positive moments of the number of cycles are infinite.

The simplest model for the recycle flow in hierarchical porous media is the one which ascribes to a given number of branches of the ultrametric structure discussed in Section 2 a cycle index. By observing that an ultrametric structure should have at least one branch, n = 1, 2, ..., and that the cycles are labeled starting from zero, m = 0, 1, 2, ..., we should have

$$m = n - 1. \tag{55}$$

We express the probability, ε_m , that *m* cycles exist in two different ways: as a ratio of the flow rate Q_m corresponding to the *m*th cycle to the total flow rate Q and in terms of the probability $\tilde{\zeta}(n)$ that an ultrametric structure is made up of *n* branches. We have:

$$\varepsilon_m = Q_m/Q = \tilde{\xi}(m+1), \tag{56}$$

where $\tilde{\xi}(n)$ is given by eq. (6). By assuming an internal backmixing mechanism, eq. (41) remains valid with the difference that the ratio Q_m/Q is given by eq. (56). By considering that $(a, b) = (a_0, b_0) = constant$ and combining eqs (6), (35), (39), (41) and (56) we obtain:

$$\psi(\tau) = \sum_{m=0}^{\infty} \sum_{q=0}^{\infty} (1-\alpha)\alpha^{q}(1-\beta)^{q}$$
$$\times [1-(1-\beta)^{q}]^{m}b^{-m}\Phi_{0}(\tau/b^{m})$$
$$= \sum_{q=0}^{\infty} (1-\alpha)\alpha^{q}\tilde{\phi}_{q}(\tau), \tag{57}$$

where

$$\tilde{\phi}_{q}(\tau) = \sum_{m=0}^{\infty} (1-\beta)^{q} [1-(1-\beta)^{q}]^{m} b^{-m} \Phi_{0}(\tau/b^{m}),$$
(58)

is an overall probability density of the residence time corresponding to the *q*th level of the ultrametric structure, that is, to the population of pores from the *q*th level.

Equations (57) and (58) give an integrated account of two different kinds of random behavior: the first one is due to the stochastic nature of the recycling process and the second one is due to the hierarchical structure of the porous medium, which is also stochastic. The model described by these equations is isomorphic with the process of stochastic amplification on ultrametric structures discussed in Section 4. By comparing eqs (21) and (22) with eqs (57) and (58) we come to the following "dictionary"

$$\tau \leftrightarrow X, \quad \tau' \leftrightarrow X', \quad m \leftrightarrow n-1,$$
 (59)

$$\psi(\tau) \leftrightarrow P^*(X), \quad \tilde{\phi}_q(\tau) \leftrightarrow \tilde{P}_q(X), \quad \phi_0(\tau') \leftrightarrow P_0(X'), \tag{60}$$

$$b_0 = b \leftrightarrow b, \quad \alpha \leftrightarrow \alpha, \quad \beta \leftrightarrow \beta. \tag{61}$$

If we assume that the probability density $\Phi_0(\tau)$ of the residence time corresponding to the 0th cycle is characterized by a cutoff value τ_c for which $\Phi_0(\tau)$ is practically equal to zero [eq. (52)] then the dictionary should be supplemented by eq. (51).

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By using the equivalence between the two models it follows that the probability densities of the residence time, $\bar{\Phi}_1(\tau), \bar{\Phi}_2(\tau), \ldots$, corresponding to the populations of pores from the different levels have long tails of the inverse power law type modulated by periodic functions of the logarithm of the residence time [eq. (23)], the corresponding fractal exponents decreasing exponentially with the label index, q[eq. (26)]. These logarithmic oscillations are due to the recycle flow; the only influence of the ultrametric structure is the exponential dependence of the fractal exponents on the level index, q. By assuming the existence of a cutoff value $\tau_{\rm c}$ of $\Phi_0(\tau)$ the asymptotic behavior of the overall probability density of the residence time is given by a very broad logarithmic tail; the corresponding proportionality factor is a superposition of oscillatory functions in $\ln (\tau/\tau_c)$ due to recycling and of much slower oscillatory terms in $\ln \left[\ln \left(\tau / \tau_c \right) \right]$ due to the hierarchical porous medium [see eqs(28)-(31)].

An attractive feature of the approaches based on the use of the probability density of residence time $\psi(\tau)$ is that this function can be measured by means of tracer experiments [15]. If we inject an inert tracer into the fluid stream entering the system then the input and output concentrations as functions of time $C_{in}(t)$, $C_{out}(t)$ are related to each other through the superposition law [15]:

$$C_{\rm out}(t) = \int C_{\rm in}(t')\psi(t-t') \, \mathrm{d}t'.$$
 (62)

In particular if the input concentration has the shape of a delta function,

$$C_{\rm in}(t) \sim \delta(t),$$
 (63)

then the output concentration is proportional to the overall probability density of the residence time,

$$C_{\rm out}(t) \sim \psi(t).$$
 (64)

For observing the logarithmic oscillations of the probability density of the residence time $\psi(\tau)$ it is tempting to analyze the answer of the system to input concentrations which are periodic functions of $\ln(t/\tau_c)$ and $\ln[\ln(t/\tau_c)]$, respectively

$$C_{\rm in} = C_{\rm in} [\ln (t/\tau_{\rm c})], \qquad (65)$$

$$C_{\rm in} = C_{\rm in} \{ \ln [\ln (t/\tau_{\rm c})] \}.$$
 (66)

By varying the periods of oscillation of the functions (65) and (66) and recording the response of the system it might be possible to evaluate the periods of oscillation in $\ln (\tau/\tau_c)$ and ln [ln (τ/τ_c)] of the tail of $\psi(\tau)$. It seems plausible to assume that the output concentration, $C_{out}(t)$, displays a kind of resonance phenomenon for these frequencies; unfortunately we have been unable to prove that this is indeed the case. The main difficulty is that in eq. (62) we have a convolution product in real time and on the other hand the oscillations of C_{in} [eqs (65) and (66)] do not take place in real time but rather in two different logarithmic scales, ln (t/τ_c) and ln [ln (t/τ_c)], respectively; the answer of the system $C_{out}(t)$ to the excitations $C_{in}(t)$ given by eqs (65) and (66) is not really a stationary frequency response. Because of this the standard form of the Fourier analysis cannot be applied for excitations described by eqs (65) and (66); in order to clarify the way in which these excitations can be used for the study of logarithmic oscillations further investigations are necessary.

7. The mechanism of logarithmic oscillations

Although the above analysis provides a mathematical description of the multiple logarithmic oscillations, it does not clarify their mechanism of generation; the physical significance of the model is hidden in the mathematical formalism.

The problem of fluid dynamics discussed in Sections 5 and 6 suggests a physical explanation for the occurrence of logarithmic oscillations. We can make an analogy with the relaxation oscillations in population dynamics generated by the time delay due to the maturation of the individuals. The recycling of the fluid in the system may be described in terms of a cascade of delayed feedback processes. This analogy is rather superficial; the main difference between the two problems is that in population dynamics the oscillations occur in real time whereas in fluid mechanics they occur in two logarithmic time scales. However, in spite of this difference, the delayed feedback gives a simple explanation for the occurrence of multiple logarithmic oscillations.

We start out by investigating the fast logarithmic oscillations with a period $\ln b$ which occur in the logarithmic time scale $\ln t$. For the *q*th level of the pore hierarchy the recycle flow is described by the overall probability density, $\tilde{\phi}_{q}(\tau)$, of the residence time given by eq. (58). By making an analogy with the renormalization group approach to critical processes [12] we rewrite eq. (58) in a self-similar form

$$\tilde{\phi}_q(\tau) = (1 - v_q)\phi_0(\tau) + \frac{v_q}{b}\,\tilde{\phi}_q(\tau/b),\tag{67}$$

where

$$v_a = 1 - (1 - \beta)^q. \tag{68}$$

Equation (67) is similar to other stochastic renormlization group equations used in the literature [1, 4-6]; in order to clarify its physical significance we rewrite it as

$$\tilde{\phi}_q(\tau) = (1 - v_q)\phi_0(\tau) + v_q \int \tilde{\phi}_q(\tau')\delta(\tau - \tau'b) \,\mathrm{d}\tau'. \tag{69}$$

The physical interpretation of eq. (69) is simple. We note that v_a can be interpreted as the probability that at qth level the process of recycling occurs. By using this interpretation of v_q the r.h.s. of eq. (69) expresses the contributions of two complementary possibilities to $\tilde{\phi}_q(\tau)$: (a) no recycling occurs with a probability $1 - v_q$ and (b) recycling occurs with a probability v_a . Equation (68) has the structure of a feedback equation: the answer of the system to recycling, $\tilde{\phi}_{q}(\tau)$, is determined by an excitation, $\tilde{\phi}_q(\tau')$, which was itself also an answer at a previous time, τ' . The delay $\tau - \tau'$ is not an additive function; due to the fact that from cycle to cycle the volume involved in the flow process is getting bigger and bigger and the flow rate is getting smaller and smaller the excitation time, τ' , is related to the answer time, τ , by a multiplicative, rather than additive law ($\tau = \tau' b$). As a result an additive delay does not exist in real time, but rather in a logarithmic time scale

$$\theta_1 = \ln \left(\tau / \tau_c \right), \tag{70}$$

where τ_c is a characteristic residence time introduced for dimensional consistency. By expressing the probability densities of the residence time in terms of θ_1 ,

$$\phi_0(\tau) d\tau = \phi_0^*(\theta_1) d\theta_1,$$

$$\phi_0^*(\theta_1) = \phi_0[\tau_c \exp(\theta_1)]\tau_c \exp(\theta_1),$$
(71)

and

$$\tilde{\phi}_{q}^{*}(\theta_{1}) = \tilde{\phi}_{q}[\tau_{c} \exp(\theta_{1})]\tau_{c} \exp(\theta_{1}), \qquad (72)$$

the renormalization group equation (67) becomes an additive delay equation

$$\tilde{\phi}_{q}^{*}(\theta_{1}) = (1 - v_{q})\phi_{0}^{*}(\theta_{1}) + v_{q}\,\tilde{\phi}_{q}^{*}(\theta_{1} - \ln b).$$
(73)

The solution of this delay equation has the following asymptotic behavior

$$\tilde{\phi}_q^*(\theta_1) \sim \exp\left(-\theta_1/\theta_1^c\right) \Pi_1(\theta_1) (\ln b)/\theta_1^c \quad \text{as } \theta_1 \to \infty,$$
 (74)

where

$$\theta_1^c = \ln b / \ln v_q \approx (1 - \beta)^{-q} \ln b, \tag{75}$$

is a characteristic logarithmic time and $\Pi(\theta_1)$ is a periodic function of θ_1 with a period ln b

$$\Pi_1(\theta_1 + \ln b) = \Pi_1(\theta_1). \tag{76}$$

The above analysis shows that the oscillations in the $\ln t$ scale are due to a delayed feedback generated by recycling at a given level: the logarithmic scale of these oscillations is generated by the multiplicative structure of the delay in real time.

The slow logarithmic oscillations with a period $-\ln (1 - \beta)$ which occur in the ln ln t scale can be interpreted in a similar way. These oscillations correspond to a delayed feedback due to the exchange of fluid among the pores from the different levels. The hierarchical structure of pores leads to a feature which is typical for an ultrametric topology [2, 3]. The minimal path connecting two close pores may be very long; due to the tree structure of the pore hierarchy such a minimal path may involve very large pores belonging to the lower levels. Taking into account that at a given level recycling may occur the delay corresponding to the whole hierarchy of pores is much longer than the delay corresponding to recycling. The multiplicative structure of the ultrametric topology leads to a logarithmic time scale in θ_1 , that is to a double logarithmic scale in the real time.

By inserting eqs (74) and (75) into eq. (57) we obtain

$$\psi^{*}(\theta_{1}) = (1 - \alpha) \sum_{q=0}^{\infty} \alpha^{q} \exp \left[-(1 - \beta)^{q} \theta_{1} / \ln b \right] \Pi_{1}(\theta_{1}) (1 - \beta)^{q},$$
(77)

where

$$\psi^*(\theta_1) = \psi[\tau_c \exp(\theta_1)]\tau_c \exp(\theta_1)$$
(78)

is the probability density of the residence time for the whole system expressed in the logarithmic time scale θ_1 . Equation (77) can be transformed into two renormalization group equations similar to eqs (67) and (69),

$$\psi^{*}(\theta_{1}) = (1 - \alpha) \exp \left(-\theta_{1}/\ln b\right) \Pi_{1}(\theta_{1})$$
$$+ \alpha (1 - \beta) \psi^{*} \left[(1 - \beta) \theta_{1}\right], \tag{79}$$
$$\psi^{*}(\theta_{1}) = (1 - \alpha) \exp \left(-\theta_{1}/\ln b\right) \Pi_{1}(\theta_{1})$$

+
$$\alpha \int \delta [\theta_1 - \theta_1'/(1-\beta)] \psi^*(\theta_1') d\theta_1'$$
. (80)

These relationships are feedback equations with a multiplicative delay; indeed the answer logarithmic time, θ_1 , is related to the excitation logarithmic time, θ'_1 , by the multiplicative law $\theta_1 = \theta'_1/(1 - \beta)$. This multiplicative delay can be transformed into an additive one by means of a second logarithmic transformation,

$$\theta_2 = \ln \theta_1 = \ln \ln (\tau/\tau_c). \tag{81}$$

By expressing the probability density of the residence time for the whole hierarchy of pores in this new time scale we have

$$\psi^{**}(\theta_2) = (1 - \alpha) \exp \left[\theta_2 - \exp \left(\theta_2\right)/\ln b\right] \Pi_1 \left[\exp \left(\theta_2\right)\right]$$
$$+ \alpha \psi^{**} \left[\theta_2 + \ln \left(1 - \beta\right)\right], \tag{82}$$

where

$$\psi^{**}(\theta_2) = \psi^{*}[\exp(\theta_2)] \exp(\theta_2)$$
$$= \psi\{\tau_c \exp[\exp(\theta_2)]\}$$
$$\times \tau_c \exp[\exp(\theta_2)] \exp(\theta_2). \tag{83}$$

The asymptotic behavior of the solution of eq. (83) is given by

$$\psi^{**}(\theta_2) \sim \exp(-\theta_2/\theta_2^c) \Pi_2[\exp(\theta_2), \theta_2] \text{ as } \theta_2 \to \infty,$$

where

$$\theta_2^c = \ln \left(1 - \beta \right) / \ln \left[\alpha (1 - \beta) \right], \tag{85}$$

(84)

and $\Pi_2(x, y)$ is a double periodic function of x and y with periods $\ln b$ and $-\ln (1 - \beta)$, respectively,

$$\Pi_2(x + \ln b, y) = \Pi_2[x, y - \ln (1 - \beta)] = \Pi_2(x, y).$$
(86)

By expressing in eq. (86) the variable θ_2 in terms of the real time we recover an asymptotic behavior for $\psi(\tau)$ similar to the one given by eq. (28).

8. Discussion

The logarithmic decay law given by eq. (28) is much slower than the inverse power laws given by eqs (8), (14) and (23): for $1 \ge H > 0$ not only all positive moments of X are infinite, but also all positive moments of $\ln (X/X_m)$ are infinite. A physical picture for this type of asymptotic behavior has been given in Sections 6 and 7 in the context of recycle flows in porous media. This picture is also valid in the general case: in comparison with Shlesinger-Hughes amplification discussed in Section 3, the process of amplification on ultrametric structures is much more efficient. By comparing eqs (12) and (13) and (20)-(26) we note that the corresponding probability distributions, ε_n and $\tilde{\xi}(n)$, are different: ε_n has an exponential shape whereas $\tilde{\xi}(n)$ has a long tail of the power law type. Besides, the probability density $\tilde{P}_q(X)$ of the random variable X corresponding to a given level q has also a long tail due to stochastic amplification. The superposition of these two long tails of $\tilde{\xi}(n)$ and $\tilde{P}_q(X)$ leads to the logarithmic decay law given by eq. (28).

Two types of logarithmic oscillations are generated by a multiplicative feedback mechanism. Their interaction leads to amplitude modulation: the logarithmic oscillations in ln (X/X_m) due to the stochastic amplification are modulated by a much slower oscillatory process in ln $[\ln (X/X_m)]$ due to the ultrametric structure. This fact outlines the "hierarchy into hierarchy" structure characteristic to our model: the amplification processes; the ultrametric structure is also hierarchic. According to eq. (21) to each level of the ultrametric hierarchy corresponds a hierarchy of amplification events described by the probability density $\tilde{P}_q(X)$.

In a recent paper (Vlad [14]) we have derived a logarithmic decay law similar to eq. (28) for the probability density of the waiting times attached to an ultrametric structure with random energy barriers. In this case a long tail due to an exponential distribution of energy barriers interacts with another long tail due to the ultrametric structure. As the long tail due to the randomness of the energy barriers is non-oscillatory no interaction of logarithmic oscillations exists.

Another possible approach related to the "hierarchy on hierarchy" structures is to construct a model with multiple stochastic amplification processes by assuming that the average number of amplification events is also a random variable subject to stochastic amplification. A repeated application of such a scheme leads to very broad probability densities whose tails are inverse powers of the multiple iterated logarithm of the random variable $\ln \cdots \ln (X/X_m)$; the main trend is modulated by a superposition of many logarithmic oscillatory terms involving the multiple iterated logarithms of the random variable $\ln (X/X_m)$, $\ln [\ln (X/X_m]]$, ..., $\ln \cdots \ln (X/X_m)$. The interaction of these multiple oscillations is much more complicated than the amplitude modulation described by eq. (31). A preliminary report on this type of stochastic amplification is presented in Ref. [29]. However, this paper does not deal with the interference of multiple logarithmic oscillations; for the sake of simplicity only a special limit is studied for which the very broad logarithmic tails are still present but the logarithmic oscillations vanish.

Concerning the hydrodynamic application presented in this paper we should say that the fluid flow in porous media has already been analyzed in connection with the possible occurrence of fractal and multifractal structures [30, 31]. However, these reports do not make use of the concept of statistical fractals and do not investigate the possible existence of multiple logarithmic oscillations of the type considered here; they focus on the fluid invasion in porous media [30] and on the occurrence of the random geometrical fractal interfaces resulting from immiscible displacements of fluids [31].

The hydrodynamic model presented in this paper is very simple. We have considered only the case of internal backmixing with constant probabilities $a = a_1 = a_2 = \cdots$ and constant effective amplification factors $b = b_1 = b_2 = \cdots$; if these two parameters are random then only the recycling in homogeneous systems without pores can be analyzed in a simple way. A general study of eq. (42) for an arbitrary probability density f(a, b) with finite moments can be done by means of a method developed by Vlad [32] in other physical contexts. The asymptotic behavor of the tail of $\Psi(\tau)$ is given by a sum of inverse power tails with different fractal exponents modulated by logarithmic oscillatory functions in ln (τ/τ_c) with different characteristic periods. We have tried, without success, to generalize this approach to recycle flows in porous media.

The problem of external backmixing is much more complicated. Even the flows in homogeneous systems cannot be analyzed exactly. For external backmixing in eq. (41) the functions $\Phi_m(\tau)$ should be replaced by the multiple time convolution products of $\Phi_0(\tau), \ldots, \Phi_m(\tau)$.

$$\Phi_m(\tau) \to \Phi_0(\tau) \otimes \Phi_1(\tau) \otimes \cdots \otimes \Phi_m(\tau). \tag{87}$$

Vlad [22] has developed an approximate method for the evaluation of the asymptotic behavior based on a double Laplace and Mellin transformation. Unfortunately this method cannot be extended for recycle flows in porous media.

9. Conclusions

In this paper we have analyzed the interaction of the logarithmic oscillations due to the occurrence of ultrametric and statistical fractal structures. The main result is that the oscillations occur in two different logarithmic scales, $\ln (X/X_m)$ and $\ln [\ln (X/X_m)]$, characteristic for the statistical fractal and ultrametric structures, respectively and the amplitude of the oscillations due to the statistical fractal structure is modulated by the oscillations due to the ultrametric structure.

As an illustration of our approach we have discussed a simplified model for the recycle flows in hierarchical porous media. The study of recycle flows has shown that the mechanism of generation of multiple logarithmic oscillations involves two types of multiplicative feedback processes. Besides, the recycle flows present the advantage that the probability density of residence time can be measured by means of tracer experiments. This example from fluid mechanics does not exhaust the possible applications of our approach. The theory developed here is of interest in connection with the study of very slow relaxation processes, occurring for instance in frozen systems far from equilibrium. Another possible application is the analysis of multiple hierarchical aggregation; the limit law (28) may describe the process of multiple hierarchical fractal aggregation corresponding to a catalytic mechanism [33].

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