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MATHEMATICAL MODELING OF ANOMALOUS DIFFUSION IN POROUS MEDIA

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Abstract. Analysis of diffusion in a complex environment shows that the conventional diffusion equation based on Fick's law fails to model the anomalous character of the diffusive mass transport observed in the field and laboratory experiments. New mathematical models of diffusive transport, different from Fick's law, were proposed and validated in literature. In the present paper the examples of the equations that can be used for describing the anomalous mass transport are presented and some important properties of these equations are discussed. Two regimes of anomalous diffusion are identified. One regime, which is called sub-diffusion, is characterized by the slower propagation of the concentration front, so that the squared distance of the front passage requires longer time than in the case of the classical Fickian diffusion. The second regime (called super-diffusion) is characterized by the higher diffusion rate, so that the particles will pass the specified distance faster than in the case of classical Fickian diffusion. Both regimes can be modeled by non-local diffusion equation with temporal and spatial fractional derivatives. It is shown that equation with spatially variable diffusivity proposed by O'Shaughnessy and Procaccia (1985), which provides a relatively good model of diffusion on a regular fractal, is less applicable for describing the effects of sub and super diffusion that may take place in a fractured porous medium or any other complex medium.

1. Introduction

The elementary particles under the effect of different force fields of different nature perform complex motion. The trajectories of these particles reproduce the geometrical objects of complex structure [2]. In this case the probability that the particles will be at the given points of space at the certain moments of time no longer can be described by the Gauss' distribution and, hence, cannot be modeled by the diffusion equation based on classical Fick's law. In the previous studies, based on the probabilistic models for migrating particles, it was shown that in complex situations the density of probability distribution can be described by equations that contain fractional spatial and temporal derivatives [7, 8, 10, 11, 23–25]. Fractured porous medium has a very complex structure, which can be considered as fractal. Hence, the material particles while migrating along fractures and pore channels will perform complex motion, imposed by the complex geometry of the pores and fractures and their distributions in the domain. So that the role of the force field in this case will be taken by the porous channels and stochastically distributed fractures. Based on the similarities of the aforementioned processes we can expect that the diffusion equation in the porous medium will be similar to the

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equations of anomalous diffusion validated for the case of elementary particles motion under the effect of different force fields.

Nigmatullin [26–28] was the first who derived fractional diffusion equation for the media of fractal geometry. Considering a comb-like structure of the medium, he obtained equation with fractional temporal derivative which models the process of “slow” diffusion (sub-diffusion). Similar approach was utilized by Fomin et al. [17] for modeling diffusion in a fractured porous medium. In the latter study, based on double porosity model [5], fractional advection diffusion equation in fractured porous aquifer was derived analytically. Expression for the coefficient in front of the fractional derivative was obtained and all parameters that can affect its value are identified. It was also shown that the order of the fractional derivative in the advection-diffusion equation depends on the fractal dimension of the porous. Application of these equations for modeling mass transport in the fractured porous medium can be found in [15, 16].

Different approaches for modeling diffusion in the media of fractal geometry were investigated extensively during the last years [29, 18, 9, 12–14]. Relatively full reviews of these approaches to this problem can be found, for example in [19, 33, 34].

In the present study we will focus on two major approaches for modeling diffusion on fractals. First approach is based on utilizing the fractional differential equations [28, 33, 34, 17] and another one is based on introduction of the variable diffusivity of the medium, reciprocal to a power function of spatial variable, whose exponent is called the index of anomalous diffusion [29].

2. Differential equations for anomalous diffusion

One-dimensional problem mass transport due to diffusion can be presented in the following equation:

$$\frac{\partial c}{\partial t} = -\frac{\partial}{\partial x}(J_c), \tag{1}$$

where J_c is the diffusive mass flux. In the case of Fickian diffusion

$$J_c = -D^{(m)}\frac{\partial c}{\partial x}, \tag{2}$$

Introducing new non-dimensional variables, $X = \frac{x}{x_0}$, $\tau = \frac{t}{t_0}$, $C = \frac{c}{c_0}$, where x_0, t_0, and c_0 are the characteristic scales, equations (1) and (2) can be presented in the following non-dimensional form:

$$\frac{1}{t_0}\frac{\partial C}{\partial \tau} = \frac{1}{x_0^2}\frac{\partial}{\partial X}\left(D^{(m)}\frac{\partial C}{\partial X}\right), \tag{3}$$

As it can be seen, equations (1) and (2) being rewritten in non-dimensional coordinates (3) preserve their original form if the spatial and temporal scales are related as $x_0 = t_0$. The latter correlation between the scales is typical for the classical Fickian diffusion. The numerous experiments with fractal objects [19] demonstrated that the above correlation for these objects does not work. In contrast, it was confirmed that the mean-square displacement of a random walker, $\langle x^2 \rangle$, depends on time as follows:

$$\langle x^2 \rangle \sim t^{2/(2+\theta)}, \tag{4}$$

where parameter θ is called the index of the anomalous diffusion. From equation (4) it immediately follows that correlations between the corresponding scales should be as

$$x_0^2 \sim t_0^{2/(2+\theta)}. \tag{5}$$

Obviously, as a particular case, the correlation for the Fickian diffusion follows from equation (5) letting in the latter $\theta = 0$. In order to obtain the adequate equation of mass balance (instead of equations (1) and (2)) for which the scales will satisfy equation (5), one has to modify the equation for the mass flux in such a way that the invariance of the diffusion equation will lead to the correlation between the scales given by equation (5). Furthermore, the expected modification of the equation for the mass flux for the certain conditions, namely, if $\theta = 0$, must resemble the classical Fick's law.

Obviously, the possible expression for the mass flux that corresponds to the correlation for scales (5) is not unique and, therefore, the diffusion equation constructed on its basis may take quite different forms.

Two possible methods marked by the letters (a) and (b), which allow us to satisfy the scaling restrictions for the anomalous diffusion, are discussed below.

Method (a) is based on assumption that the coefficient of diffusion in equation (2) is defined by the formula [29]:

$$D^{(m)}(x) = D_f x^{-\theta}, \tag{6}$$

where the effective coefficient of diffusion, D_f, is constant. It leads to the following diffusion equation:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}\left(D_f x^{-\theta}\frac{\partial c}{\partial x}\right). \tag{7}$$

It can be easily seen that for this equation the correlation (5) for the scales is satisfied. The value of the index of anomalous diffusion, θ, is determined by the fractal dimension of the medium d_f. For example, for the Koch curve [19] the correlation between θ and d_f can be readily obtained as follows. For this fractal, the triple increase of the spatial scale leads to the 16-fold increase of the time scale. Hence, we have $16 = 3^{2+\theta}$ or $\theta + 2 = 2\ln 4/\ln 3$. Recalling that fractal dimension for the Koch curve is defined as $d_f = \ln 4/\ln 3$, the coupling equation can be presented as $\theta = 2d_f - 2$. The similar reasoning for the Serpinski gasket [19] leads to the following expressions $\theta = d_f - 2 + \tilde{\zeta}$, $\tilde{\zeta} = \ln[(n+3)/(n+1)]/\ln 2$, where $\tilde{\zeta}$ characterizes the relationship between the resistivity and conductivity of the medium and n is the Euclidian dimension of the space, where the Serpinski gasket is considered; the fractal dimension in this case $d_f = \ln 3/\ln 2$.

In another method (b) it is assumed [4] that the mass flux is proportional to the fractional derivative of concentration with respect to spatial coordinate of the order $\theta + 1$. Then the correlation for the scales (5) is satisfied. However, in this case the order of the corresponding diffusion equation is grater than 2, namely, is equal to $\theta + 2$, and, therefore, it is required to have a greater number of boundary conditions than in the case of Fickian diffusion when $\theta = 0$. This fact makes doubtful the suggested representation for the mass flux. In order to satisfy the physically meaningful boundary conditions, the order of spatial derivative in the diffusion equation should be equal to 2 or less. For this purpose, in the equation for the mass flux (2) the incorporated spatial derivative should have order one or less than 1 (in the latter case it accounts for the possible spatial non-locality) and, in order to satisfy the correlation for scales (5), this spatial derivative should be accompanied by the supplemental inclusion of the effects of temporal non-locality, which can be achieved by introduction of fractional temporal derivative. The fact that variation of the mass flux with time exhibits a non-local behavior has been verified by the numerous theoretical and experimental studies [20, 25, 11]. Thus, let us consider the following expression for the mass flux:

$$J_c = D_f \partial_t^{1-\gamma}\left(\frac{\partial^\beta c}{\partial x^\beta}\right), \quad 0 < \gamma, \quad \beta < 1, \tag{8}$$

where γ and β indicate the order of the temporal and spatial fractional derivatives, respectively. According to Caputo definition, spatial and temporal fractional derivatives in equation (8) can be represented by the following expressions [31]:

$$\partial_x^\beta c = \frac{\partial^\beta c}{\partial x^\beta} = \int_0^x \frac{(x-\xi)^{-\beta}}{\Gamma(1-\beta)}\frac{\partial c}{\partial \xi}\,d\xi, \quad \partial_t^\gamma c = \frac{\partial^\gamma c}{\partial t^\gamma} = \int_0^t \frac{(t-\xi)^{-\gamma}}{\Gamma(1-\gamma)}\frac{\partial c}{\partial \xi}\,d\xi, \tag{9}$$

where $\Gamma(x)$ is Gamma function [1]. Substituting expression (8) into the equation of the mass balance (1) yields

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}\left(D_f \partial_t^{1-\gamma}\left(\frac{\partial^\beta c}{\partial x^\beta}\right)\right), \tag{10}$$

where parameters γ and β, which determine the order of the fractional derivatives, should be coupled in such way that the correlation for the scales (5) will be satisfied. The scale analysis of equation (10) leads to the following expressions relating the spatial and temporal scales:

$$t_0 = x_0^{(1+\beta)/\gamma}. \tag{11}$$

Comparing the latter expression with equation (5) yields

$$\theta + 2 = (1+\beta)/\gamma. \tag{12}$$

The analogous relationship was obtained in [35]. Equation (10) can be converted to the more symmetric form by applying to both sides of this equation the operation of fractional integration [31]. As a result, equation (10) will be converted to the following form

$$\frac{\partial^\gamma c}{\partial t^\gamma} = \frac{\partial}{\partial x}\left(D_f \frac{\partial^\beta c}{\partial x^\beta}\right). \tag{13}$$

As it was mentioned above, assuming that the porous medium has a comb-like structure of fractal geometry, Nigmatullin [26–28] obtained the same equation with $\beta = 1$

for describing diffusion in this type of porous medium. Equation (13) can be found in many publications related to the diffusion phenomenon in the chaotic migration of the particles [25, 11]. Comparing analytical solutions of equation (13) with experimental data, Fomin et al [16] demonstrated that this equation can be effectively used for modeling the anomalous contaminant diffusion from a fracture into a porous rock matrix with an alteration zone bordering the fracture.

Let us turn to derivation of the diffusion equation on fractal by utilizing the approach based on representing the diffusion coefficient as a function of the spatial coordinate (method (a)) . We will restrict ourselves to the case when concentration is the function of time and distance r from the origin to the arbitrary point in the domain within the Euclidian space, $c(t,r)$, and will assume that diffusion takes place in the direction from the origin to the arbitrary point defined by the vector $\vec{r}$. In these coordinates the diffusion equation (1) in the porous medium can be rewritten as follows:

$$\frac{\partial mc}{\partial t} = -\frac{1}{r^{n-1}}\frac{\partial}{\partial x}(r^{n-1}mJ_c), \tag{14}$$

where m is the porosity and n is the dimension of the space, so that for the one-dimensional case $n=1$, and for the axial and spherical symmetry $n=2$ and $n=3$, respectively. The porosity for the fractal medium can be obtained by the formula $m=\frac{V_p}{V_{dr}}=\frac{N\hat{V}_p}{V_{dr}}$, where N is a number of pores, $\hat{V}_p$ is the mean volume of a pore, and V_{dr} is the selected elementary layer of thickness dr. It is well known that for the fractal medium [29] the number of elements $\hat{n}$ with characteristic size r can be defined by the following equation:

$$\hat{n}(r) = a_1 r^{d_f}, \tag{15}$$

where d_f is the fractal dimension and a_1 is some constant. Utilizing equation (15), allows us to determine N and V_{dr}, so that $N=a_1 d_f r^{d_f-1}dr$ and $V_{dr}=V_{r+dr}-V_r=a_2 d_f r^{d-1}dr$. Accounting for the latter expressions, porosity m can be presented by the formula,

$$m(r) = m_* r^{d_f-n}, \tag{16}$$

where $m_*=a_1 d_f \hat{V}_p/a_2$ is a constant that characterizes the porosity. Substituting expression (16) into equation (14) gives

$$\frac{\partial c}{\partial t} = -\frac{1}{r^{d_f-1}}\frac{\partial}{\partial r}(r^{d_f-1}J_c) \tag{17}$$

where, following the results of [29], the constitutive equation for the mass flux J_c can be given by Fick's law,

$$J_c = -D^{(m)}\frac{\partial c}{\partial r}, \tag{18}$$

in which for the fractal medium, the coefficient of diffusion can be assumed to be proportional to the power function $r^{-\theta}$:

$$D^{(m)} = D_f r^{-\theta}. \tag{19}$$

Taking into account formulae (18) and (19), equation (17) can be converted to

$$\frac{\partial c}{\partial t} = \frac{D_f}{r^{d_f-1}} \frac{\partial}{\partial r} \left(r^{d_f-1-\theta} \frac{\partial c}{\partial r} \right). \tag{20}$$

Solution of equation (20) is in a good agreement with the results of exact calculations for the Serpinski gasket available in [29].

It is worth noting that in order to satisfy the correlation (5) we have to choose the proper constitutive expressions for the mass flux. Following the ideas of [29], we selected the Fick's law with the variable coefficient of diffusion which depends on the spatial variable. Taking different forms of the constitutive equation, obviously will lead to different equations. For example, as it was shown above, expressing the mass flux through the fractional derivatives, will lead to the fractional differential equation.

3. Principal properties of the equations of anomalous diffusion and their discussion

Analysis of diffusion in a complex environment (diffusion affected by the various force fields or diffusion in complex media, for example on fractals) shows that the conventional diffusion equation based on Fick's law with constant coefficient of diffusivity fails to model the anomalous character of the diffusive mass transport observed in the field and laboratory experiments. As it was mentioned above, in many complex situations, the Gaussian distribution of concentration, which is typical for the ordinary Fickian diffusion, often differs from the distributions obtained experimentally. In these situations new mathematical models of diffusive transport, different from Fick's law, were proposed and validated. The examples of the equations that can be used for describing the anomalous mass transport are given by equations (13) and (20). Some important properties of these equations will be discussed below.

3.1. Properties of the equation of diffusion

It can be readily shown that equation (20) is invariant (retains its form) regarding the following one-parametric family of transformations:

$$t' = b^{2+\theta} t, \quad r' = br, \quad c' = \mu bc \tag{21}$$

where b is a parameter and μ is some arbitrary constant.

Knowing the transformations of type (21), it is possible to obtain solutions of equation (20), which posses the same property, i.e. do not change their form after applying the transformations (21). In order to demonstrate this, let us tie the family of transformations (21) with the differential operator X_μ referred to as an infinitesimal operator [3, 21]

$$X_\mu = \xi^1 \frac{\partial}{\partial t} + \xi^2 \frac{\partial}{\partial r} + \xi^3 \frac{\partial}{\partial c}, \tag{22}$$

where coefficients ξ^1, ξ^2, and ξ^3 are defined by the following rules: $\xi^1 = \frac{\partial t'}{\partial b}\big|_{b=1}$, $\xi^2 = \frac{\partial r'}{\partial b}\big|_{b=1}$, $\xi^3 = \frac{\partial c'}{\partial b}\big|_{b=1}$ where t', r' and c' are defined by equations (21). Calculating the coefficients ξ^1, ξ^2, and ξ^3 yields

$$X_\mu = (2+\theta)t\frac{\partial}{\partial t} + r\frac{\partial}{\partial r} + \mu c\frac{\partial}{\partial c}. \tag{23}$$

Any function, which preserves its form after being subjected to transformations (21), should satisfy the following differential equation [21]:

$$X_\mu F(t,r,c) = 0, \tag{24}$$

for which the corresponding characteristic equations are rather simple:

$$\frac{dt}{(2+\theta)r} = \frac{dr}{r} = \frac{dc}{\mu c}. \tag{25}$$

The principal integrals for equations (25) can be readily found as

$$I_1 = r^{2+\theta}/t, \quad I_2 = c/t^{\mu/(2+\theta)}. \tag{26}$$

Hence, the family of transformations (21) has 2 basic invariants defined by equations (26) and, therefore, any function, which preserves its form after being subjected to transformations (21), will be the function of these two invariants, i.e.

$$F = F(I_1, I_2). \tag{27}$$

From (27) it follows that those solutions of equation (20), which do not change their form after being subjected to the transformations (21), can be given by equation $I_2 = \omega(I_1)$. Accounting for the expressions (26), the latter conclusion leads to the following representation for the solution of equation (20):

$$c(r,t) = t^{\mu/(2+\theta)}\omega(\eta), \quad \eta = r^{2+\theta}/t, \tag{28}$$

where the function $\omega(\eta)$ is a solution of the following equation, which was obtained by substituting the formulae (28) into equation (20):

$$\frac{\mu}{2+\theta}\omega - \eta\frac{d\omega}{d\eta} = \frac{D_f(2+\theta)}{\eta^{\nu-1}}\frac{d}{d\eta}\left(\eta^\nu\frac{d\omega}{d\eta}\right), \quad (\eta > 0). \tag{29}$$

After rather straightforward transformations, equation (29) can be rewritten as

$$(2+\theta)^2 D_f \eta\frac{d^2\omega}{d\eta^2} + (\nu D_f(2+\theta)^2 + \eta)\frac{d\omega}{d\eta} - \frac{\mu}{2+\theta}\omega = 0, \quad (\eta > 0) \tag{30}$$

where

$$\nu = d_f/(2+\theta). \tag{31}$$

Introducing a new variable X by the equation

$$\eta = -D_f(2+\theta)^2 X, \tag{32}$$

equation (30) can be reduced to the following form:

$$X\frac{d^2\omega}{dX^2}+(\nu-X)\frac{d\omega}{dX}-\frac{\mu}{2+\theta}\omega=0,\quad (X<0). \tag{33}$$

Equation (33) is a confluent Hypergeometric equation [1] of which the solution can be presented as

$$\omega=A_1\Phi\left(-\frac{\mu}{(2+\theta)},\nu,X\right)+A_2(-X)^{1-\nu}\Phi\left(-\frac{\mu}{(2+\theta)}-\nu+1,2-\nu,X\right),\quad (X<0) \tag{34}$$

where A_1 and A_2 are arbitrary constants and $\Phi(a,b,X)$ is a confluent Hypergeometric function [1].

Thus, expressions (28), (32) and (34) provide the family of solutions of the equation (20). Even though this family of solutions does not cover the whole spectrum of solutions of equation (20), it plays a significant role in modeling diffusion on fractals and can be effectively used for obtaining solutions of a number of boundary-value problems for equation (20). Normally, these boundary-value problems are formulated for the infinite or semi-bounded domains, because specifically such types of domains allow an introduction of the self-similar variable η. To this end, it is expedient to estimate the behavior of the obtained solution (34) for $X\to-\infty$. An asymptotic behavior of the confluent Hypergeometric function $\Phi(a,b,X)$ is well documented [1]. We will confine our analysis to the leading terms in asymptotic expansion of $\Phi(a,b,X)$. As a result, taking into account asymptotic presentation of $\Phi(a,b,X)$ [1], equation (34) reduces to

$$\omega=\left[A_1\frac{\Gamma(\nu)}{\Gamma(\nu+\mu/(2+\theta))}+A_2\frac{\Gamma(2-\nu)}{\Gamma(1+\mu/(2+\theta))}\right](-X)^{\mu/(2+\theta)}+O(|X|^{-1+\mu/(2+\theta)}), \tag{35}$$

where $\Gamma(x)$ is Gamma function. Normally, solutions that vanish as $X\to-\infty$ are of major interest. Physical this condition means that far away from the source of mass, the concentration of solute retains its initial value. Obviously, as it follows from the solution (35), $\omega\to 0$ as $X\to-\infty$ for the arbitrary μ, only if the expression in the square brackets vanishes. As a result of the latter condition, the following correlation for the constants A_1 and A_2 should be required

$$A_2=-A_1\frac{\Gamma(\nu)\Gamma(1+\mu/(2+\theta))}{\Gamma(\nu+\mu/(2+\theta))\Gamma(2-\nu)}. \tag{36}$$

Substituting expression (36) into the formula (34) and again returning to the variable η defined by (32) yields

$$\begin{aligned}\omega(\eta)=A_1\Bigg[&\Phi\left(-\frac{\mu}{(2+\theta)},\nu,-\frac{\eta}{D_f(2+\theta)}\right)-\frac{\Gamma(\nu)\Gamma(1+\mu/(2+\theta))}{\Gamma(\nu+\mu/(2+\theta))\Gamma(2-\nu)}\\ &\times\left(\frac{\eta}{D_f(2+\theta)^2}\right)^{1-\nu}\Phi\left(-\frac{\mu}{(2+\theta)}-\nu+1,2-\nu,-\frac{\eta}{D_f(2+\theta)^2}\right)\Bigg].\end{aligned} \tag{37}$$

Furthermore, due to the correlation (36), asymptotically (for the big values of η) function $\omega(\eta)$ is equivalent to the remainder term in the right-hand side of formula (35), i.e.,

$$\omega(\eta) \sim A \left(\frac{\eta}{D_f (2+\theta)^2} \right)^{\mu/(2+\theta)-1}, \tag{38}$$

where A is some constant. From the formula (38) it follows that $\omega \to 0$ as $\eta \to \infty$, only if the following restriction is imposed

$$\frac{\mu}{(2+\theta)} - 1 < 0. \tag{39}$$

Thus, expressions (28) and (37) determine solutions of equation (20) which satisfy the condition in infinity

$$r \to \infty, \quad c(r,t) \to 0, \tag{40}$$

if inequality (39) is realized. An arbitrary constant A_1 in expression (37) can be defined by accounting for the specific conditions of the concrete physical problem. As an example, let us consider the problem when the overall amount of transported mass remains constant over the entire process of diffusion, which mathematically can be modeled by imposing the following condition:

$$\int_0^\infty mc(r,t) r^{n-1}\, dr = \text{const.}, \tag{41}$$

where m is the porosity and n is the dimension of Euclidian space. Substituting m defined by expression (16), condition (41) will take the following form:

$$\int_0^\infty c(r,t) r^{d_f -1}\, dr = c_M, \tag{42}$$

where c_M is a given constant. Substitution of formula (28) into equation (42) gives the following condition for the function $\omega(\eta)$:

$$\frac{t^{(\mu+d_f)/(2+\theta)}}{2+\theta} \int_0^\infty \omega(\eta) \eta^{\nu-1}\, d\eta = c_M, \tag{43}$$

where parameter ν is defined by the formula (31). In order to satisfy the condition (43), it should be required that

$$\mu = -d_f \tag{44}$$

Substituting this value of μ into equation (37) and accounting for the fact that $\Gamma(0) = \infty$, it yields

$$\omega = A_1 \Phi \left(\nu, \nu, -\frac{\eta}{(2+\theta)^2 D_f} \right). \tag{45}$$

Using the correlation between the exponential and hypergeometric functions [1], expression (45) can be recast to

$$\omega = A_1 \exp \left(-\frac{\eta}{(2+\theta)^2 D_f} \right). \tag{46}$$

Substituting expression (46) into equation (43) and accounting for the integral representation of the Gamma function [1], the value of the parameter A_1 can be readily obtained:

$$A_1 = \frac{(2+\theta)c_M}{\Gamma(\nu)[(2+\theta)^2 D_f]^\nu}. \tag{47}$$

Combining the formulae (46) and (47), we obtain the following expression for $\omega(\eta)$:

$$\omega(\eta) = \frac{c_M(2+\theta)}{\Gamma(d_f/(2+\theta))[(2+\theta)^2 D_f]^{d_f/(2+\theta)}} \exp\left(-\frac{\eta}{(2+\theta)^2 D_f}\right). \tag{48}$$

Finally, substituting (48) into equation (28), the concentration of solute can be presented as

$$c(r,t) = \frac{c_M(2+\theta)t^{-d_f/(2+\theta)}}{\Gamma(d_f/(2+\theta))[(2+\theta)^2 D_f]^{d_f/(2+\theta)}} \exp\left(-\frac{r^{2+\theta}}{(2+\theta)^2 D_f t}\right). \tag{49}$$

The interesting property of the function $c(r,t)$ defined by equation (49) is that the limit of this function as $t \to 0$ is equal up to a factor to the Dirac delta function. In order to prove it, let us consider the limit of the following expression as $t \to 0$,

$$(f,c) = \int_0^\infty f(r)c(r,t)r^{d_f-1}\,dr. \tag{50}$$

Substituting c defined by equation (28) into the expression (50) and introducing the variable $\eta = r^{2+\theta}/t$ yields

$$\lim_{t\to 0}(f,c) = \lim_{t\to 0}\frac{1}{2+\theta}\int_0^\infty f(t^{1/(2+\theta)}\eta^{1/(2+\theta)})\omega(\eta)\eta^{\nu-1}\,d\eta. \tag{51}$$

In order to evaluate the limit of the improper integral (51) for $t \to 0$, it is important to investigate the convergence of this improper integral. Decomposing the interval of integration into two sub-intervals, $(0,N)$ and (N,∞) and estimating the value of the integral (51) taken from N to ∞, it can be readily seen that

$$\begin{aligned}\left|\int_N^\infty f(t^{1/(2+\theta)}\eta^{1/(2+\theta)})\omega(\eta)\eta^{\nu-1}\,d\eta\right| &\leqslant \int_N^\infty \left|f(t^{1/(2+\theta)}\eta^{1/(2+\theta)})\omega(\eta)\right|\eta^{\nu-1}\,d\eta \\ &\leqslant A_3\int_N^\infty \omega(\eta)\eta^{\nu-1}\,d\eta, \quad \forall t\end{aligned} \tag{52}$$

where A_3 is some constant. The inequalities (52) take place because f is bounded and $\omega(\eta)$ is a positive function. The last integral in the chain of inequalities (52) approaches zero as N approaches ∞ due to the fact that function $\omega(\eta)$ should satisfy the mass conservation condition (43). Hence, for $N \to \infty$,

$$\int_N^\infty f(t^{1/(2+\theta)}\eta^{1/(2+\theta)})\omega(\eta)\eta^{\nu-1}\,d\eta \to 0 \tag{53}$$

uniformly regarding t. Due to this uniform convergence, the order of operations of integration and taking a limit in the expression (51) can be changed. As a result of evaluating the limit under the integral sign and accounting for the formulae (43) and (44), expression (51) can be presented in the following form:

$$\lim_{t\to 0}(f,c) = \frac{f(0)}{2+\theta}\int_0^\infty \omega(\eta)\eta^{\nu-1}\,d\eta = c_M f(0). \tag{54}$$

Finally, accounting for the notation introduced in equation (50), expression (54) can be recast as

$$\lim_{t\to 0}\frac{1}{c_M}\int_0^\infty f(r)c(r,t)r^{d_f-1}\,dr = f(0). \tag{55}$$

Hence, based on the above result and referring to the definition of the Dirac delta function $\delta(r)$, it can be concluded that

$$c(r,0) = \frac{c_M}{r^{d_f-1}\delta(r)}. \tag{56}$$

Referring to this formula, solution (49) can physically interpreted as a representation of the concentration variation due to the instant injection of the solute at the point $r=0$ when at the initial moment of time the total mass of the diffusing solute c_M is accumulated at this point. The characteristic profiles of the spatial distributions of concentration on a regular fractal for different moments of time are presented in Fig. 1.

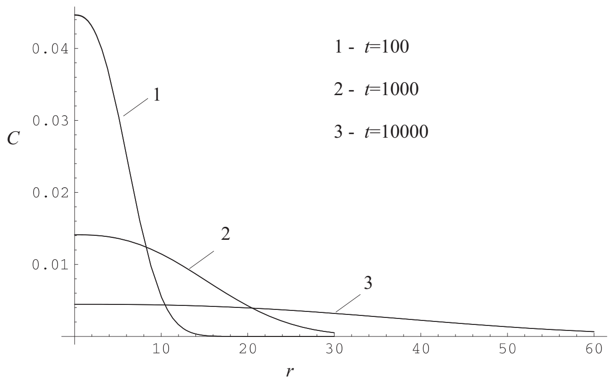

Fig. 1. Variation of the solute concentration c *on the Koch curve at the different moments of time.* $(D_f = 4D/[(2+\theta)\alpha]^2,\ D=0.4,\ \alpha=0.82-1.05,\ c_M=0.3968.)$

This figure illustrates the results of computations based on equation (49) for the Koch curve for which $d_f = \ln 4/\ln 3$. The input data, D_f and c, are taken from [14] as: $D_f = 4D/[(2+\theta)\alpha]^2$, $D = 0.4$, $\alpha = 0.82-1.05$, $c_M = 0.3968$. The results of our calculations for the Koch curve are in good agreement with calculations carried out by the authors of the above-cited paper. Furthermore, as it follows from [29, 32] solution (49) also demonstrates perfect consistency with the numerical study of random walks on the Serpinski gasket. The demonstrated capability of the obtained solution of the boundary value problem for equation (20) for modeling diffusion on the Koch curve and Serpinski gasket justifies its applicability for the other, more complex media, which can be considered as regular fractals.

It should also be noted that solution of the equation in the infinite domain $c_f(r,t)$, which satisfies the initial condition of type (56), where $c_M = 1$, in mathematical physics is referred to as a fundamental solution. If the fundamental solution is known, then solution of the equation for an arbitrary initial condition

$$t = 0, \quad c(r,t) = c_*(r), \tag{57}$$

can be readily obtained. For example, solution of equation (20) in the domain $(0,\infty)$, which satisfies the initial condition (57), can be presented as

$$c(r,t) = \int_0^\infty c_*(r) c_f(r,t) r^{d_f - 1} \, dr \tag{58}$$

where $c_f(r,t)$ is the fundamental solution defined by equation (49), where c_M is set equal to 1.

Another interesting property of the solution of equation (20) is the correlation between the mean squared distance and time. At first, let us introduce a new important quantity, $\left\langle r^{2+\theta} \right\rangle$, by the following integral:

$$\left\langle r^{2+\theta} \right\rangle = \frac{1}{c_M} \int_0^\infty r^{2+\theta} c_f(r,t) r^{d_f - 1} \, dr. \tag{59}$$

This is an important characteristic of the diffusion process that allows estimating the effective diffusivity. Apparently, the value of $\left\langle r^{2+\theta} \right\rangle$ can be calculated using $c_f(r,t)$ given by the formula (49), where c_M is set to be equal to 1. However, as it will be shown below, it is more convenient to utilize equation (20) itself rather than its solution (49). Multiplying equation (20) by $r^{(d_f-1)+(2+\theta)}/c_M$, integrating the result from 0 to ∞, and accounting for the formula (59) yields

$$\frac{d\left\langle r^{2+\theta} \right\rangle}{dt} = \frac{1}{c_M} \int_0^\infty r^{2+\theta} \frac{\partial}{\partial r} \left(D_f r^{d_f - 1 - \theta} \frac{\partial c_f}{\partial r} \right) dr. \tag{60}$$

Applying twice the method of integration by parts and accounting for the behavior of $c_f(r,t)$ at 0 and ∞, the integral in the right-hand side of equation (60) can be recast to the following form:

$$\frac{d\left\langle r^{2+\theta} \right\rangle}{dt} = \frac{(2+\theta) D_f d_f}{c_M} \int_0^\infty r^{d_f - 1} c(r,t) \, dr. \tag{61}$$

Since the solution of equation (20) satisfies the condition (42), the integral in the right-hand side of equation (61) should be equal to c_M and, therefore, equation (61) can be reduced to the following one:

$$\frac{d\left\langle r^{2+\theta} \right\rangle}{dt} = (2+\theta) D_f d_f. \tag{62}$$

The initial condition for equation (62) follows from the formulae (59) and (56):

$$t = 0, \quad \left\langle r^{2+\theta} \right\rangle = 0. \tag{63}$$

Solution of equation (62) with initial condition (63) is rather straightforward:

$$\left\langle r^{2+\theta}\right\rangle = (2+\theta)d_f D_f t. \tag{64}$$

So, if the value of $\left\langle r^{2+\theta}\right\rangle$ is a known quantity, then the effective diffusivity D_f can be readily obtained from equation (64). Moreover, the latter equation can be used for confirming the correlation between the spatial and temporal scales for diffusion (5). For example, letting in (64) $\theta = 0$ and $D_f = D_0$, which corresponds to the classical Fickian diffusion with constant diffusivity D_0, leads to the well-known correlation between the squared distance and time:

$$\left\langle r^2\right\rangle = 2d_f D_0 t, \tag{65}$$

where the values of $d_f = 1,2,3$ in this case correspond to the dimension of Euclidian space. In order to obtain the value of $\left\langle r^2\right\rangle$ for the case of anomalous diffusion with variable diffusivity we will use the obtained solution (49). Unfortunately, the direct integration of equation (20) as it was done for obtaining expression (64) does not work in this case. Following the notation of the formula (59), an expression for $\left\langle r^2\right\rangle$ can be defined as

$$\left\langle r^2\right\rangle = \frac{1}{c_M}\int_0^\infty r^2 c_f(r,t) r^{d_f-1}\,dr. \tag{66}$$

Substituting solution (49) into the integral (66), introducing there a new variable $x = r^{2+\theta}/[tD_f(2+\theta)^2]$, and accounting for the well documented properties of the Gamma function (Abramowitz and Stegun, 1972) yields

$$\left\langle r^2\right\rangle = \frac{\Gamma[(d_f+2)/(2+\theta)]}{\Gamma[d_f/(2+\theta)]}[D_f(2+\theta)]^{\frac{2}{2+\theta}} t^{\frac{2}{2+\theta}}. \tag{67}$$

Formula (67) indicates that, in contrast to the Fick's law with the constant coefficient when $\left\langle r^2\right\rangle$ and t are coupled by a linear function (65), for the anomalous diffusion the correlation between the mean squared distance and time is non-linear. Figure 2 illustrates the variation of the quantity $\left\langle r^2\right\rangle$ versus time for the regular (equation (65)) and anomalous (equation (67)) diffusion. As it can be readily seen, the time required for solute to pass the specified distance is longer for the process of anomalous diffusion than in the case of classical Fickian diffusion. Even if the diffusivity is taken several times smaller (see Fig. 2 b) the described above tendency will still become apparent starting from some finite moment of time. This type of anomalous diffusion is known as a sub-diffusion.

3.2. Properties of the equation (13) describing the anomalous non-local diffusion

Similarly to the case investigated above (equation (20)), equation (13) also preserves its form if subjected to the following family of transformations:

$$t' = b^{(\beta+1)/\gamma}t, \quad r' = br, \quad c' = \mu bc. \tag{68}$$

Duplicating the arguments of the previous subsection, it can be easily shown that solution of equation (13) can be presented in the following form:

$$c(x,t) = t^{\mu\gamma/(\beta+1)}\omega(\eta), \quad \eta = x^{(\beta+1)/\gamma}/(tD_f^{1/\gamma}). \tag{69}$$

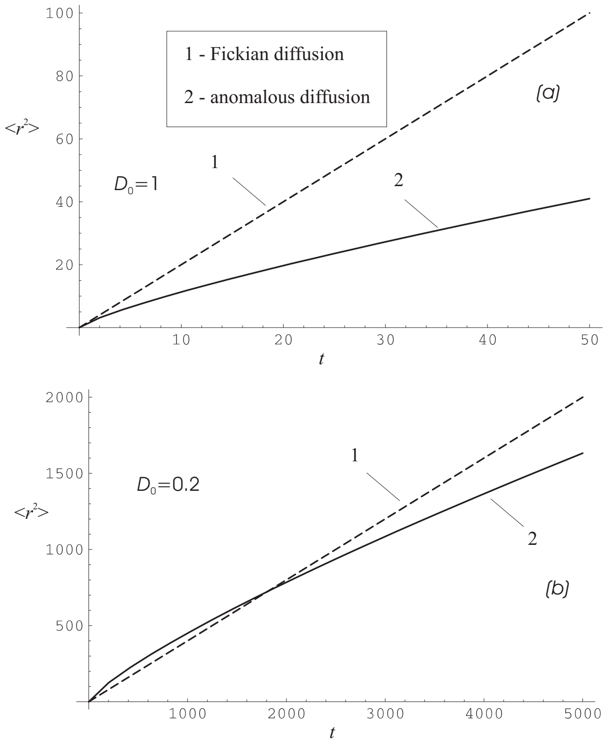

Fig. 2. The dependence of the mean square displacement $\langle r^2 \rangle$ *versus time (* $D_f = 1,\ d_f = 1.27,\ \theta = 0.5$ *).*

Accounting for the fact that for fractal type media, $(\beta+1)/\gamma = 2+\theta$, it can be seen that the form of expression (69) completely resembles the formula (28). However, in the present case, function $\omega(\eta)$ is determined by a completely different equation. This equation can be found substituting expression (69) into equation (13). Unfortunately, the resulting equation is rather awkward, so that its analytical closed-form solution cannot be guaranteed. Therefore, postponing the change of variables x and t to the variable η, let us at first apply the Laplace transform with respect to variable t for solving equation (13). Using the well-documented properties of the Laplace transform, equation (13) can be converted to the following equation in transforms

$$s^\gamma \bar{c} - s^{\gamma-1} c(x,0) = D_f \frac{\partial}{\partial x}\left(\frac{\partial^\beta \bar{c}}{\partial x^\beta}\right), \tag{70}$$

where $\bar{c}(x,s) = L[c(x,t)]$ is the Laplace transform of the function c. Let us consider the situations when either $c(x,0) = 0$ or, as in the case considered in the previous subsection, $c(x,0) = c_M \delta(x)$. Since in both former and latter cases $c(x,0) = 0$ for all

$x \neq 0$, from (70) it follows that

$$\frac{\partial}{\partial x}\left(\frac{\partial^{\beta}\bar{c}}{\partial x^{\beta}}\right) - \frac{s^{\gamma}}{D_f}\bar{c} = 0. \tag{71}$$

Without the loss of generality this equation can be considered in the domain $(0,\infty)$. (The case of negative x can be discussed analogously). In order to find the unique solution of equation (71) we need two boundary conditions. One condition follows from the fact that solution should vanish as $x \to \infty$. As in the case discussed in sub-section 3.1, this condition means that far away from the source of mass, the concentration of solute retains its initial value. The second boundary condition can be determined from the presentation of solution of equation (13) in the form of (69). Setting in (69) $x = 0$, leads to the following formula:

$$c(0,t) = A_1 t^{\mu t/(\beta+1)}, \tag{72}$$

where A_1 is some constant. Applying to $c(0,t)$ the Laplace transform, expression (72) will take the following form:

$$\bar{c}(0,s) = A_1\Gamma(1+\mu\gamma/(1+\beta))s^{-1-\mu\gamma/(\beta+1)}. \tag{73}$$

Expression (73) can serve as a second boundary condition for equation (71). The boundary value problem (A12), similar to the problem (71) and (73), is considered in Appendix A. Its solution is determined by equation (A17). Comparing the boundary-value problem (A12) with the problem (71) and (73), the solution of the latter can be easily constructed by setting in (A17) $c_0 = A_1\Gamma[1+\mu\gamma/(1+\beta)]s^{-1-\mu\gamma/(\beta+1)}$, and $\lambda = s^{\gamma}/D_f$. As a result,

$$\begin{aligned}\bar{c}(x,s) = {} & A_1\Gamma(1+\mu\gamma/(1+\beta))s^{-1-\mu\gamma/(\beta+1)}[E_{\beta+1}(s^{\gamma}x^{\beta+1}/D_f) \\ & - (s^{\gamma}x^{\beta+1}/D_f)^{\beta/(\beta+1)}E_{\beta+1,\beta+1}(s^{\gamma}x^{\beta+1}/D_f)]\end{aligned} \tag{74}$$

where $E_{\beta+1}(z)$ and $E_{\beta+1,\beta+1}(z)$ are the Mittag-Leffler functions [31]. Function $c(x,t)$ can be obtained from the equation (74) by applying the inverse Laplace transform and accounting for the Efros theorem [6]. As a result, function $c(x,t)$ can be presented in a form of a complex integral of the expressions for two particular solutions of equation (13), which can be relatively easily obtained for $\gamma = 1$, $\beta \neq 1$ and for $\gamma \neq 1$, $\beta = 1$. Unfortunately, this representation of the function $c(x,t)$ is too awkward and is not convenient for numerical calculations. In order to obtain a simple representation of the solution $c(x,t)$ it is advantageous to present the Mittag-Leffler functions in the formula (74) in a form of power series [31] and consequently apply the inverse Laplace transform to every term in these series. Thus, denoting the inverse Laplace transform by $L^{-1}[\cdot]$, expression (74) yields

$$\begin{aligned}L^{-1}[\bar{c}(x,s)] = {} & A_1\Gamma(1+\mu\gamma/(1+\beta))\{L^{-1}[s^{-1-\mu\gamma/(\beta+1)}E_{\beta+1}(s^{\gamma}x^{\beta+1}/D_f)] \\ & - L^{-1}[s^{-1-\mu\gamma/(\beta+1)}(s^{\gamma}x^{\beta+1}/D_f)^{\beta/(\beta+1)}E_{\beta+1,\beta+1}(s^{\gamma}x^{\beta+1}/D_f)]\}.\end{aligned} \tag{75}$$

Evaluating the first term in the right-hand side of equation (75), it gives

$$L^{-1}[s^{-1-\mu\gamma/(\beta+1)}E_{\beta+1}(s^{\gamma}x^{\beta+1}/D_f)] = \sum_{j=0}^{\infty}\left(\frac{x^{\beta+1}}{D_f}\right)^{j}\frac{L^{-1}[s^{\bar{\gamma}_j}]}{\Gamma[(\beta+1)j+1]}, \tag{76}$$

where $\bar{\gamma}_j = j\gamma - 1 - \mu\gamma/(1+\beta)$. The expression (76) was obtained by accounting for the definition of the Mittag-Laffer function. From the formula $L[t^{\gamma}] = s^{-1-\gamma}\Gamma(1+\gamma)$, it follows that

$$L^{-1}[s^{\gamma}] = t^{-1-\gamma}/\Gamma(-\gamma). \tag{77}$$

Due to the formula (77), expression (76) can be reduced to

$$\begin{aligned} &L^{-1}[s^{-1-\mu\gamma/(\beta+1)}E_{\beta+1}(s^{\gamma}x^{\beta+1}/D_f)] \\ &= t^{\frac{\mu\gamma}{\beta+1}}\sum_{j=0}^{\infty}\left(\frac{x^{\beta+1}}{D_f}\right)^{j}\frac{t^{-j\gamma}}{\Gamma[(\beta+1)j+1]\Gamma(1+\mu\gamma/(\beta+1)-j\gamma)}. \end{aligned} \tag{78}$$

The series in (78) determines a new function $W^{a_1,b_1}_{a_2,b_2}(z)$ defined in Appendix B, where $a_1 = 1+\mu\gamma/(\beta+1)$, $a_2 = \beta+1$, $b_1 = \gamma$, and $b_2 = 1$. In terms of these functions, expression (78) can be presented as

$$L^{-1}\left[s^{-1-\frac{\mu\gamma}{\beta+1}}E_{\beta+1}\left(\frac{s^{\gamma}x^{\beta+1}}{D_f}\right)\right] = t^{\frac{\mu\gamma}{\beta+1}}W^{1+\frac{\mu\gamma}{\beta+1},\gamma}_{\beta+1,1}\left(\frac{x^{\beta+1}}{D_f t^{\gamma}}\right). \tag{79}$$

Since the function $W^{a_1,b_1}_{a_2,b_2}(z)$ is defined only for $a_2 - b_1 > 0$, then in our case the following inequality should be satisfied, $a_2 - b_1 = 1+\beta-\gamma > 0$, where $0 < \beta$, $\gamma < 1$. Analogously to the above, the second term in the right-hand side of expression (75) can be presented as

$$\begin{aligned} &L^{-1}\left[s^{-1-\frac{(\mu-\beta)\gamma}{\beta+1}}\left(\frac{x^{\beta+1}}{D_f}\right)^{\frac{\beta}{\beta+1}}E_{\beta+1,\beta+1}\left(\frac{s^{\gamma}x^{\beta+1}}{D_f}\right)\right] \\ &= t^{\frac{\mu\gamma}{\beta+1}}\left(\frac{x^{\beta+1}}{D_f t^{\gamma}}\right)^{\frac{\beta}{\beta+1}}W^{1+\frac{(\mu-\beta)\gamma}{\beta+1},\gamma}_{\beta+1,\beta+1}\left(\frac{x^{\beta+1}}{D_f t^{\gamma}}\right). \end{aligned} \tag{80}$$

Since $c(x,t) = L^{-1}[\bar{c}(s,s)]$, correlations (75), (79) and (80) yield

$$c(x,t) = A_1 t^{\mu\gamma/(\beta+1)}[W^{1+\mu\gamma/(\beta+1),\gamma}_{\beta+1,1}(n^{\gamma}) - \eta^{\gamma\beta/(\beta+1)}W^{1+(\mu-\beta)\gamma/(\beta+1),\gamma}_{\beta+1,\beta+1}(\eta^{\gamma})] \tag{81}$$

where $\eta = x^{(\beta+1)/\gamma}/(tD_f^{1/\gamma})$ and the same letter A_1 is used for denoting the product $A_1\Gamma(1+\mu\gamma/(1+\beta))$ in expression (75). Comparing solution (81) and formulae (69), we can readily identify the appropriate expression for ω. In order to emphasize that function ω depends on parameter μ, we will write that $\omega = A\omega_{\mu}(\eta)$, where A is constant, which can be defined by the conditions imposed in the specific problem, and function $\omega_\mu(\eta)$ is given by the formula:

$$\omega_\mu(\eta) = [W_{\beta+1,1}^{1+\mu\gamma/(\beta+1),\gamma}(\eta^\gamma) - \eta^{\gamma\beta/(\beta+1)} W_{\beta+1,\beta+1}^{1+(\mu-\beta)\gamma/(\beta+1),\gamma}(\eta^\gamma)]. \tag{82}$$

Similar to the case discussed in sub-section 3.1, we can consider as an example, the diffusion problem supplemented by the condition of the total mass conservation (42). Replacing in this condition r by x gives

$$\int_0^\infty c(x,t)x^{d_f-1}\,dx = c_M. \tag{83}$$

Substituting expression (81) in (83) and accounting for the formula (82), it yields

$$A\frac{D_f^{d_f/(\beta+1)}\gamma}{(\beta+1)} t^{\frac{\gamma(\mu+d_f)}{\beta+1}} \int_0^\infty \omega_\mu(\eta)\eta^{\frac{\gamma d_f}{\beta+1}-1}\,d\eta = c_M. \tag{84}$$

In order to satisfy the condition (84), we have to set in it $\mu = -d_f$. Denoting,

$$M_{\hat{p}}^{(d_f)} = \int_0^\infty \omega_\mu(\eta)\eta^{\frac{\gamma(\hat{p}+d_f)}{\beta+1}-1}\,d\eta, \tag{85}$$

where $\hat{p}$ is some non-negative constant. Since $\gamma(\hat{p}+d_f)/(\beta+1) > 0$, the point $\eta = 0$ is not a singular point for the integral (85). Hence, the convergence of the integral (85) is determined by the convergence of its remainder,

$$\lim_{N\to\infty}\int_N^\infty \omega_\mu(\eta)\eta^{\frac{\gamma(\hat{p}+d_f)}{\beta+1}-1}\,d\eta. \tag{86}$$

Using the asymptotic representation of function $\omega_\mu(\eta)$ (see Appendix B: B12):

$$\lim_{N\to\infty}\int_N^\infty \omega_\mu(\eta)\eta^{\frac{\gamma(\hat{p}+d_f)}{\beta+1}-1}\,d\eta = \frac{(-A)D_f^\gamma}{\Gamma(-\beta)\Gamma\left[2+\frac{\mu\gamma}{\beta+1}\right]} \lim_{N\to\infty}\int_N^\infty \eta^{\gamma\left(\frac{\hat{p}+d_f}{\beta+1}-1\right)-1}. \tag{87}$$

It can be readily seen that the integral in the expression (87) converges to zero if $\gamma\left[\frac{\hat{p}+d_f}{\beta+1}-1\right] < 0$. The latter inequality imposes the restriction on the value of parameter $\hat{p}$ for which the integral (85) converges:

$$\hat{p} < \beta + 1 - d_f. \tag{88}$$

Obviously, condition (84) contains an integral (85), where parameter $\hat{p} = 0$. Therefore, due to the inequality (88) that guarantees the convergence of the integral (85), we can obtain the restriction that should be imposed on β, for which the integral (84) converges and solution of the investigated problem exists:

$$\beta > d_f - 1. \tag{89}$$

If the inequality (89) is satisfied, then constant A_1 can be obtained from equation (84) and solution of the diffusion problem for equation (13) can be presented as

$$c(x,t) = \frac{c_M(\beta+1)}{\gamma M_0^{(d_f)}} D_f^{\frac{-d_f}{\beta+1}} t^{\frac{-d_f\gamma}{\beta+1}} \omega_{-d_f}\left(\frac{x^{(\beta+1)/\gamma}}{tD_f^{1/\gamma}}\right), \tag{90}$$

where $M_0^{(d_f)}$ is defined by (85). In a similar way as for the problem discussed in subsection 3.1, in the present case it can be also shown that solution (90) of equation (13) satisfies the following condition at the initial moment of time:

$$t \to 0, \quad c(x,t) \to \frac{c_M}{x^{d_f-1}}\delta(x), \tag{91}$$

where $\delta(x)$ is the Dirac delta-function. Hence, formula (90), where c_M is set to be equal to 1, can be considered as a fundamental solution for equation (13), and, therefore, it can be used for constructing solutions when the concentration at the initial moment of time is a given function of a spatial coordinate (see, for example, solution (58) obtained for the initial condition (57)).

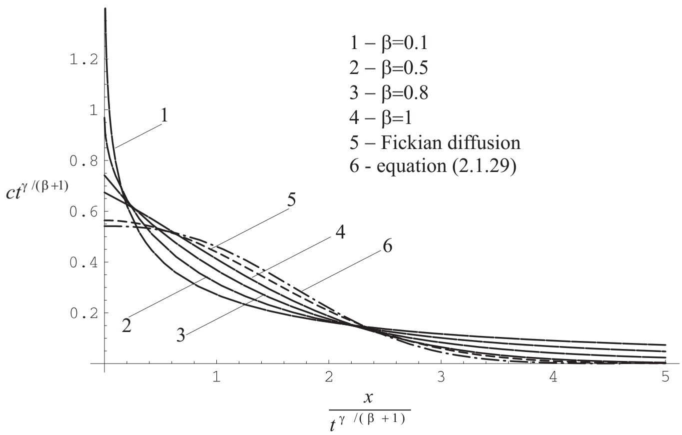

Fig. 3. Distribution of concentration in the self-similar coordinates computed by different formulae: solid lines correspond to equation (90), where $\theta = 0.5$, $D_f = 1$, $d_f = 1$, and $\gamma = (\beta+1)/(2+\theta)$; dashed line is obtained by equation (49) for $d_f = 1$ and $\theta = 0$; dot-dashed line also is obtained by equation (49) for $\theta = 0.5$.

The results of computed concentration based on solutions of equations (13) and (20) are presented in Fig. 3. The computations were carried out for the following values of controlling parameters: $d_f = 1$, $\gamma = (1+\beta)/(2+\theta)$, $\theta = 0.5$, $D_f = 1$, c_M=1, $\beta = 0.1$, 0.5, 0.8, 1. Since $d_f = 1$, then condition (89) is fulfilled for all $0 < \beta < 1$. The results presented in Fig. 3 demonstrate that the distributions of concentrations calculated by using solutions of equations (13) and (20) are substantially different. It means that equation (13) does not adequately describe the process of diffusion on a regular fractal. On the other hand, equation (20), which provides a relatively good model of diffusion on a regular fractal (this fact is discussed in sub-section 3.1), is less applicable for describing the effects of sub and super diffusion that may take place in fractured porous media [30, 22].

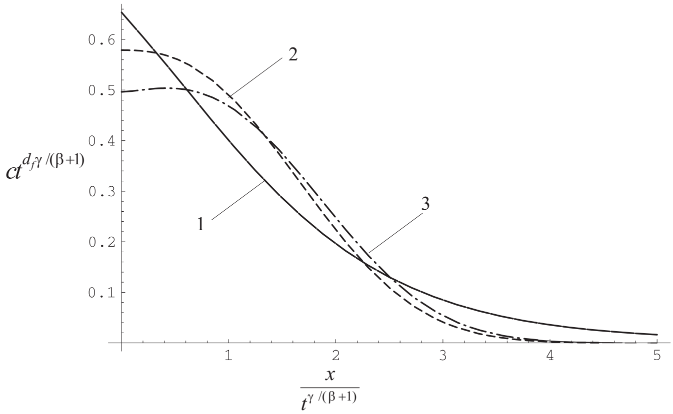

Fig. 4. Comparison of the different diffusion models for fractal medium in the case of the Koch curve. Where $\beta = 0.9,\ D_f = 1,\ \gamma = 0.7528,\ \theta = 0.5237,\ d_f = 1.27.$
(1 – equation (90); 2 – equation (49); 3 – equation (49), where for $d_f = 1$ *and* $\theta = 0$.*)*

Figure 4 confirms the above conclusion. On this figure the concentration profiles are computed for the Koch curve (see sub-section 3.1) by using different formulae. Solid curve is obtained by equation (90), dashed curve by equation (49) and the dot-dashed correspond to solution of equation (20) for $d_f = 1$ and $\theta = 0$. Since for the Koch curve $d_f \neq 1$, the value of β has to satisfy the inequality (89). Parameter β is set to be equal to 0.9. Figure 4 illustrates that if the fractal dimension is slightly bigger than 1 (same as for the Koch curve), then the simpler equation (20) can be used for modeling diffusion. Similarly to the case of equation (20) discussed in subsection 3.1, where in order to estimate the effective diffusivity of the medium the averaged distance $\left\langle r^{2+\theta} \right\rangle$ was introduced, in the present case dealing with equation (13) we can also introduce the analogous correlations:

$$\left\langle x^{\hat{p}} \right\rangle = \frac{1}{c_0} \int_0^\infty x^{\hat{p}} c(x,t) x^{d_f - 1}\, dx, \tag{92}$$

where $\hat{p}$ is some positive parameter. Substituting $c(x,t)$ given by the formula (90) into the expression (92), it yields

$$\left\langle x^{\hat{p}} \right\rangle = \frac{M_{\hat{p}}^{(d_f)}}{M_0^{(d_f)}} \left(D_f^{\frac{1}{\gamma}} t \right)^{\frac{\hat{p}\gamma}{\beta+1}}, \tag{93}$$

where $M_{\hat{p}}^{(d_f)}$ is the integral defined by the expression (85), which converges if $\hat{p}$ satisfies the inequality (88), i.e., if $0 \leqslant \hat{p} < \beta + 1 - d_f$. As it can be readily seen, the quantity $\langle x^2 \rangle$ widely used in the case of classical diffusion is not applicable for the anomalous diffusion because the condition (88) cannot be satisfied for $\hat{p} = 2$ (if, of course, $0 < \beta < 1$) and, therefore, the integral $M_{\hat{p}}^{(d_f)}$ diverges. In this situation, it is convenient to modify the definition of the mean squared distance, for instance, as

$$\{x^2\} = \lim_{\hat{p}\to 0} \langle x^{\hat{p}} \rangle^{2/\hat{p}}. \tag{94}$$

Then from (94) and (93) it follows that

$$\{x^2\} = \left(D_f^{\frac{1}{\gamma}} t\right)^{\frac{2\gamma}{\beta+1}} \lim_{\hat{p}\to 0} \left[\frac{M_{\hat{p}}^{(d_f)}}{M_0^{(d_f)}}\right]^{2/\hat{p}}. \tag{95}$$

Obviously, the expression under the limit sign depends only on parameters β, γ and d_f, and does not depend on D_f. Hence, the effective diffusivity, D_f, can be characterized by the following equation:

$$\frac{\{x^2\}}{\lim\limits_{\hat{p}\to 0} \left[\frac{M_{\hat{p}}^{(d_f)}}{M_0^{(d_f)}}\right]^{2/\hat{p}}} t^{\frac{-2\gamma}{\beta+1}} = D_f^{\frac{1}{\gamma}}. \tag{96}$$

Note that equation (95), similarly to the results of the scale analysis of equation (13), also demonstrates the temporal scale is related to the spatial scale as $x^2 \sim t^{2\gamma/(\beta+1)}$. Denoting $\lambda = 2\gamma/(\beta+1)$, equation (95) can be converted to

$$\frac{\{x^2\}}{2d_f t} = \frac{1}{2d_f} \lim_{\hat{p}\to 0} \left[\frac{M_{\hat{p}}^{(d_f)}}{M_0^{(d_f)}}\right]^{2/\hat{p}} D_f^{\frac{2}{\beta+1}} t^{\lambda-1} \tag{97}$$

where $M_{\hat{p}}^{(d_f)}$ is the integral defined by the expression (85). For the classical Fickian diffusion (with constant diffusivity and $\gamma = \beta = 1$) the left-hand side of expression (97) is constant and corresponds to the constant diffusivity. For the anomalous diffusion, which is modeled by equation (13), $\frac{\{x^2\}}{2d_f t}$ is a function of time, which variation versus time significantly depends on the value of parameter λ. Let us consider the $(\beta+1, \gamma)$-plane, where the straight line $\gamma = (1+\beta)/2$ splits the first quadrant into two zones: one where $\lambda < 1$ and the other where $\lambda > 1$. For $\lambda < 1$ the process is called sub-diffusion. Sub-diffusion is characterized by the slower propagation of the concentration front, so that the squared distance of the front passage requires longer time than in the case of the classical Fickian diffusion. For $\lambda > 1$, the process is called super-diffusion that is characterized by the higher diffusion rate, so that the particles will pass the specified distance faster than in the case of classical Fickian diffusion. The point on the line $\gamma = (1+\beta)/2$ with coordinates (2,1) corresponds to the Fickian diffusion. The other points on this line correspond to the process when the sub diffusion and super diffusion are equally developed. The variations of the function $\frac{\{x^2\}}{2d_f t}$ versus time for different regimes of diffusion are presented in Fig. 5. Since $\frac{\{x^2\}}{2d_f t}$ for the Fickian diffusion defines the diffusivity, this function can be referred to as a virtual diffusivity for the anomalous diffusion. Results presented in Fig. 5 illustrate that the process of super-diffusion takes place in the domain where $\frac{\{x^2\}}{2d_f t} > 1$, i.e. where the virtual diffusivity is greater than in Fick's law. The region in Fig. 5, where $\frac{\{x^2\}}{2d_f t} < 1$, is related to the sub-diffusion processes characterized by the lower values of the virtual diffusivity. Following this terminology, it can be readily concluded that equation (20) for all positive θ describes the process of sub-diffusion. It follows from the formula (67), where $\lambda = 2/(2+\theta) < 1$, which stipulates that $\frac{\langle r^2 \rangle}{2d_f t} \sim t^{\frac{\theta}{2+\theta}}$. Hence, equation (20) for the positive θ cannot be used for describing the process of super-diffusion.

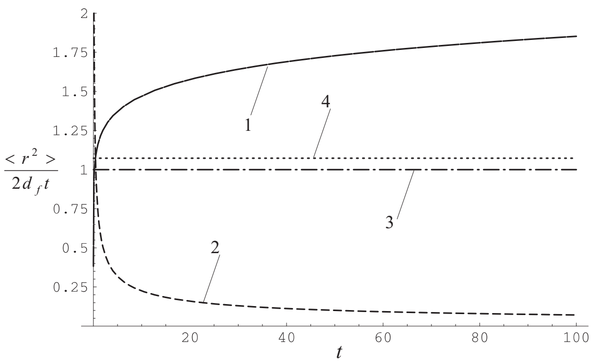

Fig. 5. Variation of the virtual diffusivity vs. time for the different regimes of diffusion. 1 – $\lambda > 1$ supper-diffusion; 2 – $\lambda < 1$ sub-diffusion; 3 – Fickian diffusion; 4 – $2\gamma = \beta + 1$.

4. Conclusions

The following conclusions can be drawn:

Two types of equations that can work well for describing the anomalous diffusion in the complex porous media, for which the spatial x_0 and temporal t_0 scales are correlated as $x_0^2 = t_0^{2/(2+\theta)}$, where θ is the constant positive parameter that depends on geometry of the porous structure.

Fick's law with the spatially variable diffusivity (so that correlation $x_0^2 = t_0^{2/(2+\theta)}$ is satisfied) leads to the conventional partial differential equation with variable coefficient, which is capable to describe diffusion on the regular fractals. Solution of this equation is found analytically by introducing the appropriate self-similar variable. Some of the interesting properties of this solution are identified. Fundamental solution for this equation is also obtained.

The time required for solute to pass the specified distance is longer for the process of anomalous diffusion (equation (67)) than in the case of classical Fickian diffusion (equation (65)). This type of anomalous diffusion is known as a sub-diffusion.

Defining the mass flux by the mixed fractional-order derivative with respect to temporal and spatial coordinates (so that correlation $x_0^2 = t_0^{2/(2+\theta)}$ is satisfied) leads to the fractional differential equation which is proven to be a good model for a number of mass transport processes in nature and industry. For example, using this fractional order diffusion equation, the non-Fickian anomalous diffusion in the porous rocks with altered region bordering the fracture is modeled and justified in [16]. In the present paper solution of this equation is found by the Laplace transform and expression for the fundamental solution is also obtained.

The computed results demonstrate that the distributions of concentrations calculated by using solutions of fractional equation (13) and equation (20) with a spatially variable diffusivity are substantially different. It means that equation (13) does not adequately describe the process of diffusion on a regular fractal. On the other hand, equation (20), which provides a relatively good model of diffusion on a regular fractal, is less applicable for describing the effects of sub and super diffusion that may take place in fractured porous media [30, 22, 16].

Appendix A. Mittag-Leffler function

Mittag-Leffler functions are defined by the following series, which are valid for the entire complex plane,

$$E_\alpha(z) \equiv \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\alpha n+1)}, \quad \alpha > 0, \quad z \in C, \tag{A1}$$

$$E_{\alpha,\beta}(z) \equiv \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\alpha n+\beta)}, \quad \alpha > 0, \quad \beta > 0, \quad z \in C, \tag{A2}$$

where C is a complex plane.

Some properties of the Mittag-Leffler function

$$E_{\alpha,\beta}(z) = 1/\Gamma(\beta) + zE_{\alpha,\beta+1}(z); \tag{A3}$$

$$E_{\alpha,\beta}(z) = \beta E_{\alpha,\beta+1}(z) + \alpha z E'_{\alpha,\beta+1}(z); \tag{A4}$$

$$\left(\frac{d}{dz}\right)^m [z^{\beta-1} E_{\alpha,\beta}(z^\alpha)] = z^{\beta-m-1} E_{\alpha,\beta-m}(z^\alpha). \tag{A5}$$

Asymptotic expansions

$$E_{\alpha,\beta}(z) = \frac{z^{(1-\beta)/\alpha}}{\alpha}\exp(z^{1/\alpha}) - \sum_{k=1}^{\infty}\frac{z^{-k}}{\Gamma(\beta-k\alpha)},\ |z|\to\infty,\ |\arg z| < \alpha\pi/2, \qquad 0<\alpha<2$$
$$E_{\alpha,\beta}(z) = -\sum_{k=1}^{\infty}\frac{z^{-k}}{\Gamma(\beta-k\alpha)}, \quad |z|\to\infty, \quad \alpha\pi/2 < \arg z < 2\pi-\alpha\pi/2. \tag{A6}$$

The Laplace transform of the Mittag-Leffler functions can be evaluated by utilizing the following relationship:

$$\int_0^{\infty}\exp(-u)E_\alpha(u^\alpha z)du = \frac{1}{1-z} = \int_0^{\infty}\exp(-u)u^{\beta-1}E_{\alpha,\beta}(u^\alpha z)du, \quad \alpha,\ \beta>0. \tag{A7}$$

From the formula (A7) it is easy to ascertain that

$$\begin{aligned} &L[E_\alpha(-\lambda t^\alpha)] = s^{\alpha-1}/(s^\alpha+\lambda), \quad \mathrm{Re}\, s > |\lambda|^{1/\alpha},\\ &L[t^{\beta-1}E_{\alpha,\beta}(-\lambda t^\alpha)] = s^{\alpha-\beta}/(s^\alpha+\lambda), \quad \mathrm{Re}\, s > |\lambda|^{1/\alpha}. \end{aligned} \tag{A8}$$

Some particular Mittag-Leffler functions

$$\begin{aligned} &E_0(z) = 1/(1-z), \quad |z|<1,\\ &E_2(z^2) = \cosh z, \quad E_2(-z^2) = \cos z, \quad z\in C\\ &E_{1/2}(\pm z^{1/2}) = \exp(z)\,\mathrm{erfc}\,(\mp z^{1/2}), \quad z\in C \end{aligned} \tag{A9}$$

where

$$\mathrm{erfc}\,(z) = 1-\mathrm{erf}\,(z), \quad \mathrm{erf}\,(z) = \frac{2}{\sqrt{\pi}}\int_0^z \exp(-u^2)\,du. \tag{A10}$$

$$\begin{aligned} &E_{1,2}(z) = [\exp(z)-1]/z;\\ &E_{2,2}(z) = \sinh(\sqrt{z})/\sqrt{z}. \end{aligned} \tag{A11}$$

Application of Mittag-Leffler functions and Laplace transform to solution of the boundary-value problem for the equation with the fractional derivative

$$\frac{d}{dx}\left(\frac{d^\beta c}{dx^\beta}\right) - \lambda c = 0; \quad x=0, \quad c=c_0; \quad x\to\infty, \quad c\to 0; \quad 0<\beta<1, \tag{A12}$$

where $\frac{d^\beta c}{dx^\beta}$ is the fractional derivative of the order β. After the Laplace transform with respect to the variable x, the differential equation in (A12) takes the form

$$s[s^{\beta}\,\bar{c} - s^{\beta-1}c_0] - q_0 - \lambda\,\bar{c} = 0, \tag{A13}$$

where $\bar{c} = L[c]$ is the Laplace transform of function c and $q_0 = \left.\frac{d^{\beta}c}{dx^{\beta}}\right|_{x=0}$, where q_0 is the unknown constant.

From (A13)

$$\bar{c} = c_0\frac{s^{\beta}}{s^{\beta+1}-\lambda} + q_0\frac{1}{s^{\beta+1}-\lambda}. \tag{A14}$$

Accounting for the formulae (A9), expression (A14) can be recast to

$$c(x,\lambda) = c_0E_{\beta+1}(\lambda x^{\beta+1}) + q_0x^{\beta}E_{\beta+1,\beta+1}(\lambda x^{\beta+1}), \tag{A15}$$

where the properties (A4) and (A5) were used for proving that

$$\lambda x^{\beta}E_{\beta+1,\beta+1}(\lambda x^{\beta+1}) = \frac{d}{dx}[E_{\beta+1}(\lambda x^{\beta+1})]. \tag{A16}$$

The formula (C15) defines the general solution for the differential equation given in (A12). In order to find the solution, which satisfies the condition in infinity, $c \to 0$, $x \to \infty$, the asymptotic formulae for the Mittag-Leffler functions (A6) can be applied to the equation (A15):

$$\lim_{x\to\infty} c(x,\lambda) = (c_0 + q_0\lambda^{-\beta/(1+\beta)})\lim_{x\to\infty}[(\beta+1)^{-1}\exp(\lambda^{1/(1+\beta)}x)] = 0.$$

The last condition is satisfied if the factor $(c_0 + q_0\lambda^{-\beta/(1+\beta)})$ is equal to zero. Therefore, $q_0 = -c_0\lambda^{\beta/(1+\beta)}$, and solution (A15) take the following form:

$$c(x,\lambda) = c_0[E_{\beta+1}(\lambda x^{\beta+1}) - \lambda^{\beta/(1+\beta)}x^{\beta}E_{\beta+1,\beta+1}(\lambda x^{\beta+1})]. \tag{A17}$$

Thus, the formula (A17) presents the solution of the problem (A12).

Appendix B. Definition of the function $W_{a_2,b_2}^{a_1,b_1}(z)$

Let consider the series

$$W_{a_2,b_2}^{a_1,b_1}(z) = \sum_{k=0}^{\infty}\frac{z^k}{\Gamma(a_2k+b_2)\Gamma(a_1-b_1k)}, \quad a_2,\ b_2,\ b_1 > 0. \tag{B1}$$

Since

$$\Gamma(z+1) = \sqrt{2\pi}|z|^{z+1/2}\exp(-z)[1+O(|z|^{-1})], \tag{B2}$$

we can write:

$$\begin{aligned}&|\Gamma(a_2k+b_2)\Gamma(a_1-b_1k)|\\ &\quad\sim(\sqrt{2\pi})^{2-a_2+b_1}\left(a_2^{a_2}b_1^{-b_1}\right)^k a_2^{b_2-1/2}b_1^{a_1-1/2}(k!)^{a_2-b_1}k^{b_2+a_1-1-(a_2-b_1)/2}.\end{aligned} \tag{B3}$$

Substituting the formula (B3) to the series (B1), it can be readily seen that the series (B1) converges absolutely along with the series:

$$\sum_{k=0}^{\infty} u_k = \sum_{k=0}^{\infty} \frac{|\hat{z}|^k}{(k!)^{a_2-b_1} k^{\chi}}, \tag{B4}$$

where $\hat{z} = (z b_1^{b_1}/a_2^{a_2})$, $\chi = b_2 + a_1 - 1 - (a_2 - b_1)/2$.

Furthermore, it can easily be shown for the terms of the series (B4) that

$$k\left(\frac{u_k}{u_{k+1}} - 1\right) \sim \frac{k^{1+a_2-b_1}}{|\hat{z}|}\left(1 - |\hat{z}|\, k^{-(a_2-b_1)}\right), \quad k \to \infty. \tag{B5}$$

Accounting for the asymptotic expression (B5), it can be concluded that, if

$$(a_2 - b_1) > 0, \tag{B6}$$

then for any positive constant A, there exists the number N such that the following inequality takes place:

$$k\left(\frac{u_k}{u_{k+1}} - 1\right) \geqslant A > 1, \quad (\forall \hat{z} < \infty, \quad k > N). \tag{B7}$$

Therefore, according to the Raabe's test (Arfken, 1985), the series (B4) and (B1) converge for any finite z. If $(a_2 - b_1) < 0$, the inequality (B7) is not satisfied and the series (B4) diverges. Therefore, the series (B1) does not converge absolutely. If $a_2 = b_1$, the inequality is satisfied only when $|\hat{z}| < 1$. In this case, the series (B4) converges when $|\hat{z}| < 1$ and, therefore, the series (B1) converges absolutely only for $|z| < 1$.

Thus, we can conclude that, if $a_2 > b_1$, the series (B1) converges absolutely and the function $W_{a_2,b_2}^{a_1,b_1}(z)$ is defined.

Asymptotic behavior of the function $\omega_\mu(\eta)$

Let us define the function $\omega_\mu(\eta)$ by the following expression:

$$\omega_\mu(\eta) = A\left[W_{\beta+1}^{1+\mu\gamma/(\beta+1),\gamma}\left(\frac{\eta^\gamma}{D_f}\right) - \left(\frac{\eta^\gamma}{D_f}\right)^{\beta/(\beta+1)} W_{\beta+1,\beta+1}^{1+(\mu-\beta)\gamma/(\beta+1),\gamma}\left(\frac{\eta^\gamma}{D_f}\right)\right] \tag{B8}$$

where A, D_f are constants and $\eta = x^{(\beta+1)/\gamma}/t$.

According to the well documented properties of Laplace transform,

$$L[t^{\frac{\mu\gamma}{\beta+1}}\omega_\mu(\eta)] = A s^{-1-\frac{\mu\gamma}{\beta+1}}\left[E_{\beta+1}\left(\frac{s^\gamma x^{\beta+1}}{D_f}\right) - \left(\frac{s^\gamma x^{\beta+1}}{D_f}\right)^{\frac{\beta}{\beta+1}} E_{\beta+1,\beta+1}\left(\frac{s^\gamma x^{\beta+1}}{D_f}\right)\right] \tag{B9}$$

where L is the Laplace transform with respect to t, and $E_a(z)$, $E_{a,b}(z)$ are Mittag-Leffler functions defined by (A1) and (A2).

Using (A5), expression (B9) can be transformed to the following form:

$$L[t^{\mu\gamma/(\beta+1)}\omega_\mu(\eta)] = As^{-1-\mu\gamma/(\beta+1)}\left[-\sum_{k=1}^{\infty}\left(\frac{s^\gamma x^{\beta+1}}{D_f}\right)^{-k}\frac{1}{\Gamma[1-(\beta+1)k]} + \left(\frac{s^\gamma x^{\beta+1}}{D_f}\right)^{\beta/(\beta+1)}\sum_{k=2}^{\infty}\left(\frac{s^\gamma x^{\beta+1}}{D_f}\right)^{-k}\frac{1}{\Gamma[(\beta+1)(1-k)]}\right]. \tag{B10}$$

After the inverse Laplace transform the latter expression gives

$$t^{\frac{\mu\gamma}{\beta+1}}\omega_\mu(\eta) = At^{\frac{\mu\gamma}{\beta+1}}\left[-\sum_{k=1}^{\infty}\left(\frac{x^{\beta+1}}{D_f t^\gamma}\right)^{-k}\frac{1}{\Gamma[1-(\beta+1)k]\Gamma[1+\gamma+\frac{\mu\gamma}{\beta+1}]} + \left(\frac{x^{\beta+1}}{D_f t^\gamma}\right)^{\beta/(\beta+1)}\sum_{k=2}^{\infty}\left(\frac{x^{\beta+1}}{D_f t^\gamma}\right)^{-k}\frac{1}{\Gamma[(\beta+1)(1-k)]\Gamma[1+\mu\gamma/(\beta+1)+\gamma]}\right]. \tag{B11}$$

Introducing here the variable $\eta = x^{(\beta+1)/\gamma}/t$, leads to the following result for $\eta\to\infty$:

$$\omega_\mu(\eta) = A\left(\frac{\eta^\gamma}{D_f}\right)^{-1}\left[\frac{(-1)}{\Gamma(-\beta)\Gamma[1+\mu\gamma/(\beta+1)+\gamma]} + \left(\frac{\eta^\gamma}{D_f}\right)^{-1/(\beta+1)}\frac{-1}{\Gamma(-\beta-1)\Gamma[1+\mu\gamma/(\beta+1)+\gamma]} + O\left(\left(\frac{\eta^\gamma}{D_f}\right)^{-1}\right)\right]. \tag{B12}$$

Since the series (B8) that defines $\omega_\mu(\eta)$ exhibits slow convergence for the big values of the variable $\eta = x^{(\beta+1)/\gamma}/t$, the formula (B12) can be readily used in this case for computing $\omega_\mu(\eta)$.

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