Solute Transport with Non-Equilibrium Adsorption in a Non-Homogeneous Porous Medium

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Abstract: - In this paper, a solute transport problem with non-equilibrium adsorption in a non-homogeneous porous medium consisting of two zones, one with high permeability (mobile zone) and another one with low permeability (immobile liquid zone) are considered. In the mobile zone, there are two zones in both of which adsorption of solute with reversible kinetics occurs. The results of this approach are compared with known, traditional approaches. It is shown that this method of modeling the process gives a satisfactory result. By appropriate selection of the parameters of the source term, one can obtain results close to those of the well-known bicontinuum approach.

Key-Words: - Adsorption, approximation, fractional derivative, porous media, retardation factor, mobile zone.

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1 Introduction

Aquifers, oil, and gas reservoirs, as a rule, have a heterogeneous structure at the micro- and macroscale, [1]. Heterogeneous reservoirs on a macro scale consist of different zones with different, sometimes very strong, filtration-capacitive properties, i.e. porosity, permeability, etc. Zones with well porosity and permeability are well conductors for liquids and various substances suspended or dissolved in fluids. A typical example of heterogeneous formations is fractured porous media (FPM), [2], [3], the structure of which is represented as a system of fractures surrounded by porous blocks.

Colloidal particles suspended in a liquid can move relatively fast and travel longer distances in structured porous media than in media with a homogeneous structure, [8], [12], [14], [15], [39]. The reason for this is the presence of pathways conducive to the fast movement of substances. In the simulation of solute transport in FPM, it is usually assumed that the main ways for moving liquid and suspended solids (or dissolved substances) are fractures. Porous blocks in simplified models are considered impermeable to liquids, but particles or solutes can penetrate into them due to the diffusion phenomenon. Thus, two zones are formed in the medium, one with a mobile fluid (fractures) and the other with an immobile one (porous blocks). Between zones, mass transfer processes occur. The advanced solute transport in a porous medium can be the result of many factors. Therefore, there are certain difficulties in the mathematical modeling of this phenomenon. Some models in this direction were presented in [10], [17], [18], [19]. The two-zone approach noted above was used in these models. Mass transfer between zones is modeled by a first-order kinetic equation, [9], [20]. A slightly different approach combining kinetic and linear mass transfer between zones was proposed in [13]. A certain modification of the two-zone approach is an approach that takes into account fluid motion in both zones, but with different scales, [10], [17].

When colloidal particles are transported in a porous medium, they are usually deposited in pores, the causes of which are varied. Depending on the nature and location of the interaction of particles with the surface of the rock skeleton, deposition can be reversible or irreversible. Given these factors, transport models naturally become more complex. Solute transport in double porosity media taking into account reversible and irreversible deposition is described by such complex models. At the same time, it is important to take into account the texture of the medium in the models, [6],7], [21]. Mass transfer between two flow zones is considered as a function of the deposited volume of the solute in each zone, in addition, small pores may be excluded from the transport process, i.e. their locking due to the deposition of substances, [6], [11], [16].

In [4] a transport model of colloidal substances in a medium with double porosity is presented, taking into account reversible and irreversible particle retention, as well as first-order mass transfer between fractures and porous blocks. The obtained analytical solution was used to describe experimental results, [5]. A good agreement was obtained between theoretical and experimental results. Dispersion and retention parameters were higher for larger particles; the intensity of reversible and irreversible particle retention was higher for a medium with relatively small pores.

In [13] a transport model in a medium with double porosity was considered taking into account the reversible and irreversible deposition of colloid particles in both zones and the first-order equilibrium mass exchange between the zones. In each zona, i.e. in fractures and porous blocks, a reversible and irreversible deposition of particles with various characteristics occurs, described by linear equations. An analytical solution to the problem is obtained, which is used to describe the results of previous experiments, [6]. Coefficients of mathematical models are defined as the solution to the coefficient inverse problems (CIP), known as identification problems, [26]. It is assumed that the coefficients of the equation depend on the spatial coordinates and are independent of time. The statements of the problems are based on the use of uniqueness theorems for the solution of the CIP proved in [25], [29], [30], [33]. To obtain a unique solution of the CIP, it is required to set an overdetermined set of boundary conditions on the boundary of the zone: the function for which the equation is written or its normal derivative.

Coefficient inverse problems (identification problems) have become the subject of intensive study, especially in recent years. Interest in them is caused primarily by their important applications. They find applications in solving problems of designing oil reservoir development (determining the filtration parameters of reservoirs), [28], [30], [32], [34], [35], [36], solving environmental monitoring problems, etc. The standard CIP statement contains a residual function, which depends on the solution of the corresponding problem of mathematical physics, [34]. Methods for the numerical solution of CIP in connection with their applications in underground hydrodynamics were developed in [25], [26], [27], [29], [31].

In this paper, an inhomogeneous two-zone medium is considered a single-zone medium with a source (sink). The second zone is modeled through the source (sink). This approach is fundamentally new because, in fact, the bicontinual medium is presented as a mono-continual one. The validity of this approach is justified by the convergence of the results on the basis of the mono-continuous approach to the corresponding results of the bicontinuous approach. In the work, this is done by minimizing the residual function. In addition, it is assumed that in both parts of the first zone there is reversible adsorption of particles with the corresponding kinetic equations. Identification of parameters in the source (sink) term in the mass balance equation is carried out by solving the corresponding CIP using data from [4].

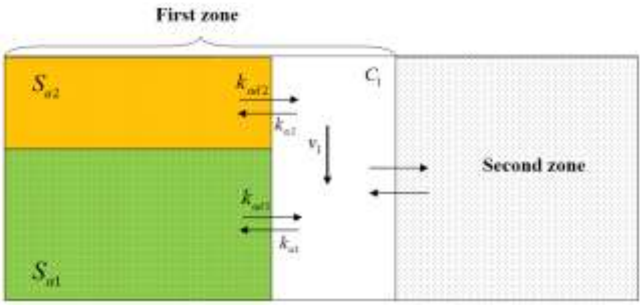


Fig. 1: Scheme of solute transport in a two-zone medium

2 The Mathematical Model and Its Numerical Implementation

An inhomogeneous porous medium is considered, consisting of well-permeable and relatively low-permeable zones, the diagram of which is shown in Fig. 1. The parameters in the first zone are indicated by index 1. There are two sections in zone 1, in each of which the particle deposition with reversible nonequilibrium nonlinear kinetics occurs. It is believed that such processes also occur in the second zone, but we will not write equations and conditions for it. With the second zone, there is an exchange of substance, which we model by the fractional-order derivative in time of the solute concentration in the first zone. Consequently, in contrast to [4], the concentration field in the second zone is not considered. Note, that the fractional approach was previously used in [37], [38], [40].

The equations of solute transport in one-dimensional case are written as

$$\rho \frac{\partial S_{a1}}{\partial t} + \rho \frac{\partial S_{a2}}{\partial t} + \theta_1 \frac{\partial C_1}{\partial t} + a_2 \frac{\partial^\gamma C_1}{\partial t^\gamma} = \\ = \theta_1 D_1 \frac{\partial^2 C_1}{\partial x^2} - \theta_1 v_1 \frac{\partial C_1}{\partial x}, \tag{1}$$

where t is time, *s, x* is distance, m, D_1 is longitudinal dispersion coefficient, m^2/s, v_1 is the fluid velocity, m/s, C_1 is volume concentration of the solute in the fluid, S_{a1} and S_{a2} are concentrations of deposited particles, m^3/kg, θ_1 is porosity, $\mathrm{m}^3/\mathrm{m}^3$, ρ is medium density, kg/m^3, a_2 *is* retardation factor related to the mass exchange between two zones, $s^{\gamma-1}$, γ is the order time derivative with respect to time, $0<\gamma\le 1$.

The deposition of particles in each of the sections of the first zone is reversible with the different kinetic equations

$$\rho\frac{\partial S_{a1}}{\partial t}=\theta_1 k_{a1}C_1-\rho k_{ad1}S_{a1}, \tag{2}$$

$$\rho\frac{\partial S_{a2}}{\partial t}=\theta_1 k_{a2}C_1-\rho k_{ad2}S_{a2}, \tag{3}$$

where k_{a1}, k_{a2} are coefficients of solute deposition from the fluid phase to the solid phase, s^{-1}, k_{ad1}, k_{ad2} are coefficients of substance detachment from the solid phase and transition into liquid, s^{-1}.

Let a fluid with a constant solute concentration c_0 be pumped into the medium initially saturated with pure (without particles) liquid from the initial moment of time. Let us consider such time periods where the concentration field does not reach the right boundary of the medium, $x=\infty$. Under the noted assumptions, the initial and boundary conditions for the problem have the form

$$C_1(0,x)=0,\ S_{a1}(0,x)=0,\ S_{a2}(0,x)=0, \tag{4}$$

$$C_1(t,0)=c_0, \tag{5}$$

$$\frac{\partial C_1}{\partial x}(t,\infty)=0. \tag{6}$$

The problem (1) - (6) although linear, obtaining an analytical solution is difficult because three concentration fields must be found at the same time. Therefore, to solve the problem, we use the finite difference method. In the considered region $\Omega=\{(t,x),\ 0\le t\le T,\ 0\le x\le\infty\}$ a uniform grid was introduced

$$\bar{\omega}_{\tau h}=\left\{\begin{matrix}(t_j,x_i);\ t_j=\tau j,\ x_i=ih,\\ \tau=\dfrac{T}{J},\ i=\overline{0,I},\ \ j=\overline{0,J}\end{matrix}\right\},$$

where I is a sufficiently large integer chosen so that segment $[0,x_I]$, $x_i=ih$, overlaps the area of the calculated change in the fields C_1, S_{a1}, and S_{a2}. h is the grid step in the x direction.

In the open grid area

$$\omega_{\tau h}=\left\{\begin{matrix}(t_j,x_i);\ t_j=\tau j,\ x_i=ih,\\ \tau=\dfrac{T}{J},\ j=\overline{1,J},\ \ i=\overline{1,I-1},\end{matrix}\right\}$$

equations (1), (2), (3) were approximated as follows

$$\rho\frac{(S_{a1})_i^{j+1}-(S_{a1})_i^j}{\tau}+\rho\frac{(S_{a2})_i^{j+1}-(S_{a2})_i^j}{\tau}+$$
$$+\theta_1\frac{(C_1)_i^{j+1}-(C_1)_i^j}{\tau}+\frac{a_2}{\Gamma(2-\gamma)}\times$$
$$\times\left[\sum_{k=0}^{j-1}\frac{(C_1)_i^{k+1}-(C_1)_i^k}{\tau}((j-k+1)^{1-\gamma}-(j-k)^{1-\gamma})+\frac{((C_1)_i^{j+1}-(C_1)_i^j)\tau^{1-\gamma}}{\tau}\right]= \tag{7}$$
$$=\theta_1 D_1\frac{(C_1)_{i-1}^{j+1}-2(C_1)_i^{j+1}+(C_1)_{i+1}^{j+1}}{h^2}-$$
$$-\theta_1 v_1\frac{(C_1)_i^{j+1}-(C_1)_{i-1}^{j+1}}{h},$$

$$\rho\frac{(S_{a1})_i^{j+1}-(S_{a1})_i^j}{\tau}=\theta_1 k_{a1}(C_1)_i^j-\rho k_{ad1}(S_{a1})_i^{j+1}, \tag{8}$$

$$\rho\frac{(S_{a2})_i^{j+1}-(S_{a2})_i^j}{\tau}=\theta_1 k_{a2}(C_1)_i^j-\rho k_{ad2}(S_{a2})_i^{j+1}, \tag{9}$$

where $(C_1)_i^j$, $(S_{a1})_i^j$, $(S_{a2})_i^j$ are grid values of functions $C_1(t,x)$, $S_{a1}(t,x)$, $S_{a2}(t,x)$ at a given point (t_j,x_i).

From the explicit grid equations (8), (9) we determine $(S_{a1})_i^{j+1}$, $(S_{a2})_i^{j+1}$

$$(S_{a1})_i^{j+1} = p_{b1}(S_{a1})_i^j + p_{b2}, \quad (10)$$

$$(S_{a2})_i^{j+1} = q_{b1}(S_{a2})_i^j + q_{b2}, \quad (11)$$

where

$$p_{b1} = \frac{1}{1+\tau k_{ad1}}, \; p_{b2} = \frac{\tau\theta_1 k_{a1}}{\rho+\rho\tau k_{ad1}}(C_1)_i^j,$$
$$q_{b1} = \frac{1}{1+\tau k_{ad2}}, \; q_{b2} = \frac{\tau\theta_1 k_{a2}}{\rho+\rho\tau k_{ad2}}(C_1)_i^j.$$

The grid equations (7) are reduced to the form

$$A_1(C_1)_{i-1}^{j+1} - B_1(C_1)_i^{j+1} + E_1(C_1)_{i+1}^{j+1} = -(F_1)_i^j, \quad (12)$$

where

$$A_1 = \frac{\theta_1 D_1 \tau}{h^2} + \frac{\theta_1 v_1 \tau}{h},$$
$$B_1 = \theta_1 + \frac{2\theta_1 D_1 \tau}{h^2} + \frac{\theta_1 v_1 \tau}{h} + \frac{a_2 \tau^{1-\gamma}}{\Gamma(2-\gamma)}$$
$$E_1 = \frac{\theta_1 D_1 \tau}{h^2},$$
$$(F_1)_i^j = (\theta_1 + \frac{a_2\tau^{1-\gamma}}{\Gamma(2-\beta)})(C_1)_i^j - \rho((S_{a1})_i^{j+1} -$$
$$-(S_{a1})_i^j) - \rho((S_{a2})_i^{j+1} - (S_{a2})_i^j) - \frac{a_2}{\Gamma(2-\gamma)} \times$$
$$\times \left[\begin{array}{l} \sum_{k=0}^{j-1}((j-k+1)^{1-\gamma} - (j-k)^{1-\gamma})(C_1)_i^{k+1} - \\ -((j-k+1)^{1-\gamma} - (j-k)^{1-\gamma})(C_1)_i^k \end{array} \right]$$

The following procedure of computing is used. From (10), (11) $(S_{a1})_i^{j+1}$, $(S_{a2})_i^{j+1}$ are determined, then we solve the system of linear equations (12) by Thomas' algorithm in order to calculate $(C_1)_i^{j+1}$. Since $p_{b1}, q_{b1} < 1$, schemes (10), (11) are stable, and for (12) the stability conditions of the Thomas' algorithm are satisfied.

To assess the performance of the proposed model, it is important to compare the results with the corresponding results, [4]. To do this, we compare the source (stock) terms $\alpha(C_2 - C_1)$ in [4] and $a_2 \frac{\partial^\gamma C_1}{\partial t^\gamma}$ in (1). To quantify the proximity of the results based on the curves was calculated

$$\delta_1 = \int_0^L (I_1 - I_2)^2 dx \quad (13)$$

for a given value of t, where L is the conditional boundary of the medium to which the concentration profiles extend,

$$I_1 = \alpha(C_2 - C_1), \quad I_2 = a_2 \frac{\partial^\gamma C_1}{\partial t^\gamma}.$$

The proximity of the terms I_1 and I_2 should guarantee the proximity of the concentration fields C_1 determined using the proposed approach and the model, [4]. To estimate their proximity, we use the standard deviation (13), only for C_1 determined on the basis of two models, i.e.

$$\delta_2 = \int_0^L \left(C_1^{(1)} - C_1^{(2)}\right)^2 dx,$$

where $C_1^{(1)}$– concentration field $C_1(t,x)$ for a given t, determined according to [4], and $C_1^{(2)}$ – the same as defined here.

For other moments t and α, a_2, γ different estimates can be obtained for δ_1 and δ_2. In principle, to approximate the two models, it is important to set and solve the corresponding coefficient inverse problems by determining of a_2, γ for a given value of α or, conversely, determining of α for a given α and γ.

3. Numerical Results and Their Analysis

In the calculations following initial values of parameters are used:

$c_0 = 0$, $v_1 = 10^{-4}\,\mathrm{m}/s$, $D_1 = v_1 \cdot \alpha_l$, $\alpha_l = 0{,}005$ m, $\rho = 1800\, kg/m^3$, $\theta_1 = 0{,}1$, $k_{a1} = 3\cdot 10^{-4} s^{-1}$, $k_{ad1} = 2{,}5\cdot 10^{-4} s^{-1}$, $k_{a2} = 4\cdot 10^{-4} s^{-1}$, $k_{ad2} = 2\cdot 10^{-4} s^{-1}$ and various a_2, γ.

Some results are shown in Fig.2. As can be seen from the figure, the outflow of particles into the second zone leads to a slow distribution of the solute concentration profiles in the mobile fluid. As a consequence of this phenomenon, delays are also observed in the concentrations of the adsorbed mass. From this, it is clear that with a decrease in the index of the fractional derivative γ from 1, both in the solute concentration in the fluid and in the concentration of the adsorbed solute in the well-permeable zone, there is a delay in the distribution.

For a certain set of parameters a_2, γ and $\alpha = 10^{-4}\,\text{s}^{-1}$, graphs I_1, I_2 are shown in Fig.3. As can be seen from the figure, the patterns of change in stock terms are similar, which indicates a qualitative agreement between the results of the proposed model and the results of the model, [4].

After that, we minimize the functional

$$\Phi(a_2,\gamma) = \int_0^T\int_0^L (I_1 - I_2)^2\,dxdt \quad , \qquad (14)$$

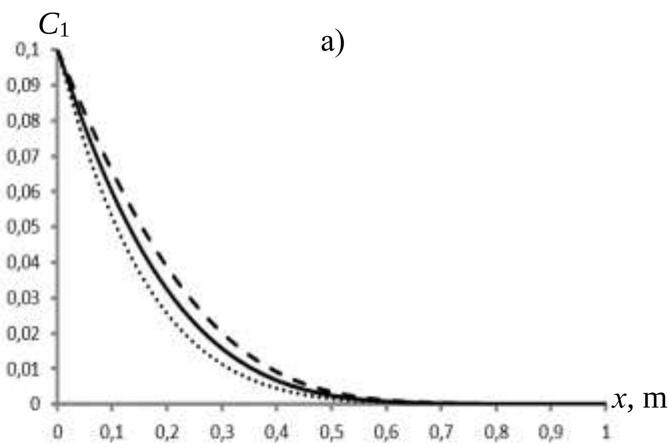
that characterizes the standard deviation of I_1 from I_2 for the entire time period. The calculations show that the minimum value of $\Phi(a_2,\gamma)$ is achieved at $a_2 = 0{,}0006,\ \gamma = 0{,}8$.

The proximity of the terms I_1 and I_2 should guarantee the proximity of the concentration fields C_1, determined using the proposed approach and model, [4]. For this, the corresponding profiles are plotted for the data, obtained through minimization of $\Phi(a_2,\gamma)$ (Fig. 4). As can be seen from the graphs, the solutions are close to each other.

For a numerical estimation of their proximity, we use the standard deviation of the type (14), only for the one determined on the basis of two models, i.e.

$$F(a_2,\gamma) = \int_0^T\int_0^L \left(C_1^{(1)} - C_1^{(2)}\right)^2 dxdt,$$

where $C_1^{(1)}$ is the concentration field $C_1(t,x)$ for a given t, determined according to [4]. $C_1^{(2)}$ is the same as defined here. For the cases analyzed above, the following minimum value of $F(a_2,\gamma)$ was obtained 0,002347387654452 for $a_2 = 0{,}0006,\ \gamma = 0{,}8$.



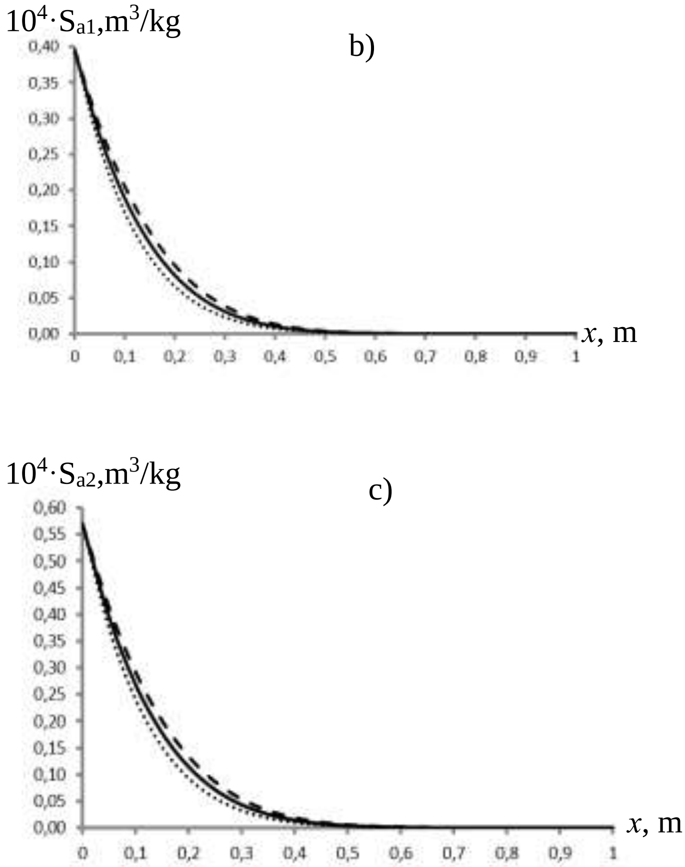
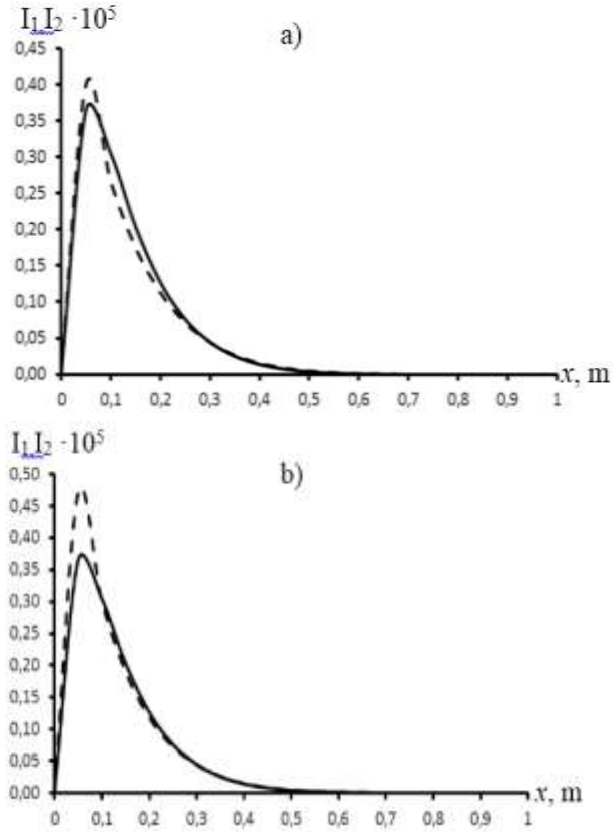


Fig. 2: Concentration profiles C_1 (a), S_{a1} (b), S_{a2} (c) at t=3600 s,
$\gamma = 0{,}5$ (⋯⋯), 0,7(——), 0,9(– – – –).



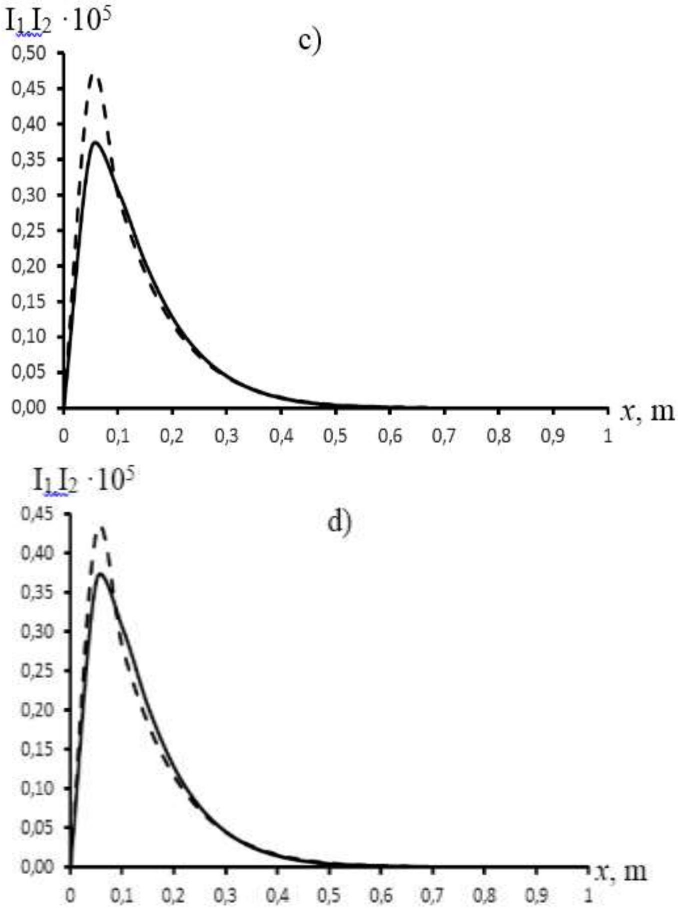


Fig. 3: Comparison of source members
$a_2 = 0{,}0001,\ \gamma = 0{,}3$ (a), $a_2 = 0{,}0002,\ \gamma = 0{,}5$ (b),
$a_2 = 0{,}0004,\ \gamma = 0{,}7$ (c), $a_2 = 0{,}0006,\ \gamma = 0{,}8$ (d),
I_1 (———), I_2 (– – – –).

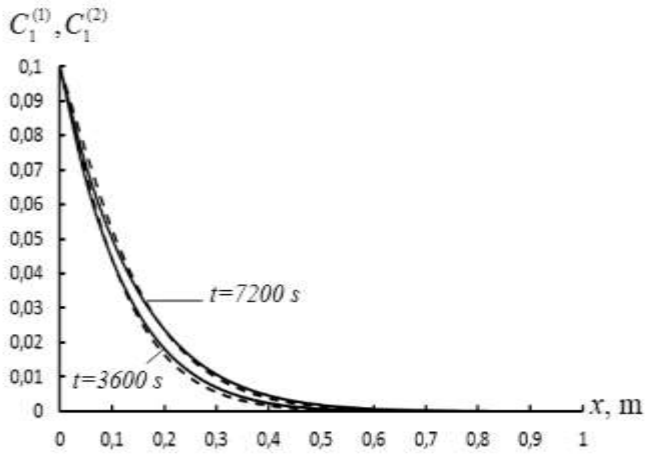


Fig. 4: Comparison of concentration profiles obtained on the basis of two models $a_2 = 0{,}0006$, $\gamma = 0{,}8$,
$C_1^{(1)}$ (———), $C_1^{(2)}$ (- - - - - -).

The analysis shows that the simpler model proposed here, with an appropriate choice of parameters, can satisfactorily describe the results of a more complex model, [4].

4 Conclusions

In contrast to [4], a new model is proposed where the presence of the second zone of an inhomogeneous medium is taken into account in the form of a source (sink) term in the transport equation written out for the first zone. The stock term is presented as a fractional time derivative of the concentration of the substance in the first zone with a certain coefficient. Thus, this approach is mono-continuous, while the bicontinual approach was used in [4]. The model was implemented numerically and the effect of mass transfer to the second zone on the transport characteristics in the first zone was estimated. It is observed that with a decrease of the order in the fractional derivative γ from 1, both in the concentration of the particles in the mobile fluid and of the adsorbed substances in the mobile zone, the dynamics of distribution delay. A problem of approximation of the results according to the proposed model with the corresponding results, [4] was solved. For this, values of parameters in the stock term, which ensures close results, are obtained using a variational approach that minimizes the residual function. It is shown that for certain values of parameters a_2 and γ a good convergence can be achieved. Thus, the fundamental possibility of the proposed mono-continuum model to describe the results of the corresponding bicontinuum model is shown. In addition, as it is shown in [22], the heterogeneity of porous media can be a cause of anomalous phenomena in filtration and transport processes. It is known, [23], [24], fractional time in filtration and transport laws can model anomalous phenomena. Therefore one can expect that the proposed here model can be used to study anomalous transport phenomena in non-homogeneous porous media.

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