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MODELING ANOMALOUS SOLUTE TRANSPORT IN FRACTURED-POROUS MEDIUM WITH NONEQUILIBRIUM ADSORPTION

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Abstract The solute transport processes in fractured porous media are characterized by complex inhomogeneities and transport mechanisms of the medium and are of significant scientific and practical importance in many engineering and environmental problems. Fractured porous media in theoretical investigations is considered as a system of fractures and adjacent porous blocks (matrix). In such media, solute transport occurs mainly through the fracture system with mass transfer in porous blocks. In this paper a problem of anomalous solute transport in an element of fractured porous media (FPM), that consists of single fracture and surrounded porous block, is studied, where non-equilibrium adsorption phenomena are taken into account. To describe solute transport a mathematical model is developed, where transport is governed by convection, diffusion, adsorption phenomena in the fracture, while only adsorption phenomena and diffusion in the porous block. Adsorption kinetics in the porous block and the fracture are different, that can be explained by their different specific surfaces. Anomalous phenomena are described by fractional derivatives in governing equations, that are adopted in Caputo sense. The problem has been solved using the finite difference method. Influence of anomality on solute transport characteristics is analysed. The results were obtained at various values of the fractional derivative orders and the adsorption coefficients considerably. The results show that at bigger values of the adsorption coefficient, the distribution of solute concentration in porous block is delayed. With adsorption the distribution of the solute in the fracture has also slowed down. With a decrease in the order of the fractional derivatives with respect to time from 1, it has been found that the distribution of the solute slows down even more that is slow diffusion.

Keywords: Anomalous solute transport, Fractional advection-dispersion, Fractional order, Nonequilibrium adsorption, Numerical solution.

1. Introduction

The processes of solute transport and filtration in fractured porous media are complex and important topics at the intersection of many scientific research fields. The specific porous structure of these media and the network of fractures make the movement of liquid and gas, as well as filtration processes, unique. For a deeper analysis of these issues, studies often take into account geophysical, hydrodynamic and geochemical parameters.

Filtration processes and fluid flows in fractures can be cited as the main mechanisms of solute transport. Darcy's law is used to explain many filtration processes, but the full operation of this law in fractured porous media may not always work correctly due to heterogeneities created by fractures [1]. The width, length, shape, and interrelationships of the fracture system directly affect filtration processes and often result in the formation of uncharacteristic flow profiles. This can dramatically change the permeability of the medium [2].

A number of special equations are used in the modeling of solute transport processes. Among these equations, Laplace, Hele-Shaw and Navier-Stokes equations play an important role. In particular, the Navier-Stokes equations are used to determine fluid dynamics and analyze flows with relatively high Reynolds numbers in fractured porous media [3]. Also, when modeling multiphase flows, for example, in oil, water and gas mixtures, it is necessary to take

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into account factors such as capillary forces and surface tension [4].

Diffusion and convection processes are seen as the main mechanisms of solute transport in fractured porous media. Diffusion processes play an important role in the movement of fluid or gas through pores and fractures, especially in the presence of solute concentration gradients [5]. At the same time, convective flows are generally considered as fluid flows driven by a pressure gradient, which increases the rate of mass transfer and leads to solute transport on a larger scale than diffusion processes [6]. In the analysis of filtration and solute transport processes, the specific dynamic properties of fractures should also be taken into account. For example, the degree of interconnection between fractures, their orientation and width significantly affect the efficiency of filtration processes [7].

The process of diffusion in the filtration, flow of particles adsorbed on the walls of channels was studied earlier in connection with the needs of the chemical industry, and it was shown that the adsorption of such particles on a real granular adsorbent is described by differential equations [8,9].

Many scientific researchers have focused on solving the problems of contaminant transport along a discrete fracture in a fractured porous medium [10–12]. In [13] the fluid flow and solute transport are modeled using a three-dimensional discrete fracture matrix system, taking into account different values of fracture density and dimensions. It was established that with an increase in the fracture density or the minimum fracture radius, the corresponding fluid flow and transport channels of the solute increase, and the range of distribution of the solute concentration in the matrix expands. In the most paper on this topic, an analytical solution is developed for solute transport in a fracture under the assumption that dispersion and diffusion along the fracture are negligible [14,15]. In [10-15] Fick's law was used for mathematical modeling of solute transport process in a fractured porous medium. Investigations conducted in recent years show that fractured porous media with a complex structure have fractal properties, and classical Fick's law cannot adequately describe the processes of solute transport in such media.

The research works [16-18] make it clear that the best way to describe the anomalous types processes of diffusion is to consider the so-called fractional kinetic equations, where the fractional derivative operator with respect to time is applied to the solute transport. It can be proved that the temporal fractional derivative is an invaluable tool, particularly for modelling and analysing reactive solute transport [19]. The papers [20-22] demonstrate the effectiveness of fractional derivatives equations in describing solute transport in porous rocks. The theoretical results align closely with laboratory and field experiments, confirming the reliability of this approach.

Numerical solution methods such as Finite Difference Method and Finite Element Method are often used to solve problems of solute transport in fractured porous media [23–25]. These methods are used to more accurately analyze the processes of solute transport. Also, experimental methods and field studies are of great importance, because they allow to confirm the results of modeling and to better understand the real conditions in fractured porous media [26].

In [27] solute transport process in a system consisting of a vertical fracture and a porous block was investigated using fractional differential equations by neglecting the adsorption phenomenon. However, it is known that when solute flows through a fractured porous medium, a portion of the solute is adsorbed on the walls of the medium, and this phenomenon significantly affects the process.

In the reviewed literature, systems consisting of a single fracture and a porous block matrix adjacent to it [28-30] were studied based on traditional models. In contrast, in this work, the mathematical model of solute transport process is improved using fractional derivatives taking into account complex structure of the medium. In addition, solute is adsorbed on the medium's walls by a non-equilibrium adsorption. The models of solute transport in the porous block and the fracture, taking into account solute transport through common boundary of two zones, are written separately using fractional differential equations. Adsorption of solute is nonequilibrium in both zones.

2. Statement of the Problem

Mathematical modeling of solute transport processes in fractured-porous media is required, simultaneously taking into account the effects of anomalous transport phenomena and non-equilibrium adsorption. Anomalous solute transport occurs in a fractured porous medium, which is consisting of two parts. The scheme of this medium is depicted in Figure 1, where the horizontally oriented fracture is called $\Omega_1 = \{0 \le x < +\infty,\, 0 \le t < \infty\}$ and porous block the adjacent to it is called $\Omega_2 = \{0 \le x < +\infty, 0 \le y < +\infty, 0 \le t < +\infty\}$ zone. A solute transport problem is posed and studied in the first quarter of the coordinate plane. That is, the x-axis is oriented along the fracture and its origin is at the point $x=0$. The Ω_1 zone is considered to be a one-dimensional object, that is, it depends only on x. In the Ω_2 zone, x and y are depended, but the mass transfer by diffusion only along the vertical y-axis. In the initial case, the entire zones are considered to be clean (without solute), that is, filled with pure water.

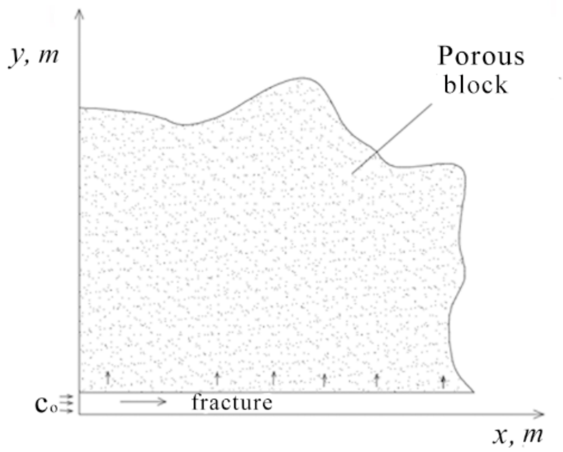


Figure 1: Pattern of fractured-porous medium

The solute transport equations have the following form:

$$\frac{\partial^\alpha c_f}{\partial t^\alpha}+\rho\frac{\partial^\alpha s_f}{\partial t^\alpha}+v\frac{\partial c_f}{\partial x}= = D_f\frac{\partial^\beta c_f}{\partial x^\beta}+m_0 D_m\frac{\partial^{1-\gamma}}{\partial t^{1-\gamma}}\left(\frac{\partial^\delta c_m}{\partial y^\delta}\right)\Bigg|_{y=0} \quad (1)$$

$$\frac{\partial^\gamma c_m}{\partial t^\gamma}+\rho\frac{\partial^\gamma s_m}{\partial t^\gamma}=D_m\frac{\partial^{1+\delta}c_m}{\partial y^{1+\delta}} \quad (2)$$

where ρ – density of the medium, kg/m^3 $c_f=c_f(t,x)$, $c_m=c_m(t,x,y)$ are solute concentration in the fracture and porous block, respectively, m^3/m^3; D_f is the coefficient of diffusion in the fracture, m^β/s^α; D_m is the coefficient of diffusion in the matrix, $m^{1+\delta}/s^\gamma$; m_0 is the matrix porosity, v is anomalous velocity of flow, m^β/s^α.

The incoming solute concentration into meduim is adsorbed in both zones. The process of adsorption is non-equilibrium, so we use the following nonequilibrium adsorption kinetics

$$\frac{\partial^\alpha s_f}{\partial t^\alpha}=\alpha_f\left(k_f c_f-s_f\right), \quad (3)$$

$$\frac{\partial^\gamma s_m}{\partial t^\gamma}=\alpha_m\left(k_m c_m-s_m\right), \quad (4)$$

where s_f and s_m are volumes of adsorbed solute per unit mass of medium, in the fracture and porous block, respectively, m^3/kg, k_m, k_f are adsorption coefficients in the matrix and in the fracture, which are physical properties of solute and rock surface, m^3/kg; α_m, α_f are the adsorption rate coefficients characterise the intensity of adsorption processes in the matrix and in fractures, respectively, $s^{-\alpha}$.

The boundary and Initial conditions are taken as:

$$c_f(0,x)=0,\quad c_m(0,x,y)=0, \quad (5)$$

$$c_f(t,0)=c_0,\quad c_f(t,\infty)=0, \quad (6)$$

$$c_m(t,x,0)=c_f(t,x),\quad c_m(t,x,\infty)=0, \quad (7)$$

$$s_m(0,x,y)=0,\quad s_f(0,x)=0. \quad (8)$$

3. Numerical Solution

The problem (1) – (8) is solved numerically using the finite difference method [31]. To approximate (1) – (8), we introduce a grid to the Ω_1 and Ω_2,

$$\overline{\omega}_{h_1h_2\tau}=\overline{\omega}^1_{h_1\tau}\cup\overline{\omega}^2_{h_1\tau},$$

$$\overline{\omega}^{\,1}_{h_1\tau}=\overline{\omega}^{\,1}_{h_1\tau}\cup\overline{\omega}^{\,1}_{h_1\tau}=\{(t_j,x_i),\ t_j=\tau j,\ x_i=ih_1,$$

$$\overline{\omega}^{\,1}_{h_1\tau}=\{(t_j,x_i),\ t_j=\tau j,\ x_i=ih_1,\quad j=\overline{0,J},\ i=0,1,\ldots,I,\ \tau=T/J\},$$

$$\overline{\omega}^{\,2}_{h_1\tau}=\{(t_j,x_i,y_k),\ t_j=\tau j,\ x_i=ih_1, y_i=kh_2,$$

$$j=\overline{0,J},\ i=0,1,\ldots,I,\ k=0,1,\ldots,K,\ \tau=T/J\},$$

where h_1, h_2 are steps by axes x, y, τ is the step by time, T is maximum time, I,K,J are sufficiently large integer numbers.

Equations (1), (2) on the introduced grid are approximated as follows:

$$\begin{aligned}
&\frac{1}{\Gamma(2-\alpha)\tau^\alpha}\left[\sum_{l=0}^{j-2}\left((c_f)_i^{p+1}-(c_f)_i^p\right)\times\right.\\
&\left.\times\left((j-p+1)^\alpha-(j-p)^\alpha\right)+(c_f)_i^{j+1}-(c_f)_i^j\right]\\
&+\frac{\rho}{\Gamma(2-\alpha)\tau^\alpha}\left[\sum_{l=0}^{j-2}\left((s_f)_i^{p+1}-(s_f)_i^p\right)\times\right.\\
&\left.\times\left((j-p+1)^\alpha-(j-p)^\alpha\right)+(s_f)_i^{j+1}-(s_f)_i^j\right]+\\
&+v\frac{(c_f)_{i+1}^j-(c_f)_i^j}{h_1}=\frac{m_0D_m}{\Gamma(2-\delta)h_2^\delta\Gamma(1+\gamma)\tau^{1-\gamma}}\times\\
&\times\left[\sum_{l=0}^{j-1}\left[\left((c_m)_{i0}^{l+1}-(c_m)_{i0}^l-(c_m)_{i1}^{l+1}+(c_m)_{i1}^l\right)\times\right.\right.\\
&\left.\left.\left(-(j-l-1)^{1-\gamma}+(j-l)^{1-\gamma}\right)\right]\right]+\frac{D_f\Gamma(2-\alpha)\tau^\alpha}{\Gamma(3-\beta)h_1^{\ \beta}}\\
&\times\sum_{q=0}^{k-1}\left((c_f)_{i-(q-1)}^j-2(c_f)_{i-q}^j+(c_f)_{i-(q+1)}^i\right)\times\\
&\times\left((q+1)^{2-\beta}-q^{2-\beta}\right),
\end{aligned} \quad (9)$$

$$\frac{1}{\Gamma(2-\gamma)\tau^{\gamma}}\left[\sum_{l=0}^{j-2}\left((c_m)_{ik}^{l+1}-(c_m)_{ik}^{l}\right)\times \times\left(-(j-l)^{\gamma}+(j-l+1)^{\gamma}\right)+(c_m)_{ik}^{j+1}-(c_m)_{ik}^{j}\right]+ + \frac{\rho}{\Gamma(2-\alpha)\tau^{\alpha}}\left[\sum_{l=0}^{j-2}\left((s_m)_i^{p+1}-(s_m)_i^{p}\right)\cdot + \left((j-p+1)^{\alpha}-(j-p)^{\alpha}\right)+(s_m)_i^{j+1}-(s_m)_i^{j}\right]= = \frac{D_m}{\Gamma(2-\delta)h_2^{1+\delta}}\times \times\sum_{r=0}^{k-1}\left((c_m)_{ik-(r-1)}^{j}-2(c_m)_{ik-r}^{j}+(c_m)_{ik-(r+1)}^{i}\right) \times\left((r+1)^{1-\delta}-r^{1-\delta}\right). \tag{10}$$

and also (3), (4)

$$\frac{1}{\Gamma(2-\alpha)\tau^{\alpha}}\left[\sum_{l=0}^{j-2}\left((s_f)_i^{p+1}-(s_f)_i^{p}\right)\times \times\left((j-p+1)^{\alpha}-(j-p)^{\alpha}\right)+ +(s_f)_i^{j+1}-(s_f)_i^{j}\right]=\alpha_f\left(k_f(c_f)_i^{j}-(s_f)_i^{j}\right), \tag{11}$$

$$\frac{1}{\Gamma(2-\alpha)\tau^{\alpha}}\left[\sum_{l=0}^{j-2}\left((s_m)_{ik}^{p+1}-(s_m)_{ik}^{p}\right)\cdot \left((j-p+1)^{\alpha}-(j-p)^{\alpha}\right)+ +(s_m)_{ik}^{j+1}-(s_m)_{ik}^{j}\right]=\alpha_m\left(k_m(c_m)_{ik}^{j}-(s_m)_{ik}^{j}\right), \tag{12}$$

Fractional derivatives in (9) – (12) are approximated as in [32, 33].

(5)-(8) conditions are approximated as:

$$(c_f)_i^0=0, \tag{13}$$

$$(c_m)_{ik}^0=0, \tag{14}$$

$$(c_f)_0^j=c_0, \tag{15}$$

$$(c_m)_{i0}^j=(c_f)_i^j, \tag{16}$$

$$(c_f)_I^j=0, \tag{17}$$

$$(c_m)_{iK}^j=0. \tag{18}$$

$$(s_f)_i^0=0 \tag{19}$$

$$(s_m)_{ik}^0=0 \tag{20}$$

Values of $(s_f)_i^j$, $(s_m)_{ik}^j$ and $(c_f)_i^j,(c_m)_{ik}^j$ are determined using (9) - (12), and (13) - (20) respectively.

4. Results and Discussion

The concentration profiles, profiles of adsorbed solute concentration are analysed using numerical results. Table 1 shows the comparative study of concentration of the fluid at section $x=0.1\ m$ and $x=0.5\ m$. In the solute transport equation, as a particular case all orders of the derivatives are fixed as integer number to verify the present results with the result ([34]). Table 1 shows that the comparative study of concentration profile. From this study, it is concluded the current analysis gives better results and is accurate.

Table 1. Comparative study of concentration profile.

	$x=0.1\ m$		$x=0.5\ m$	
y (m)	c_m/c_0 ([34] result)	c_m/c_0 (present result)	c_m/c_0 ([34] result)	c_m/c_0 (present result)
0.0	0.8564	0.813	0.3535	0.33857
0.1	0.5512	0.5215	0.2452	0.2361
0.2	0.3107	0.2999	0.1525	0.1458
0.3	0.1758	0.1681	0.0925	0.0856
0.4	0.0757	0.05	0.0415	0.0392
0.5	0.0215	0.019	0.0102	0.00992

Table 2 shows the comparative study of numerical values of adsorption concentration at distances $x=0.1\ m$ and $x=0.5\ m$. Here, we can also see that the current results almost coincide with the results of [34]. We can observe from Table 2 the change in percentage between present and available result is 6%. Therefore, in particular case with integer order derivatives the results reduce to known results.

Table 2. Comparative study of adsorbed solute concentration profile.

	$x=0.1\ m$		$x=0.5\ m$	
y (m)	$10^{-7}s_m$ ([34] result)	$10^{-7}s_m$ (present result)	$10^{-7}s_m$ ([34] result)	$10^{-7}s_m$ (present result)
0.0	0.9519	0.9284	0.4552	0.4358
0.1	0.5408	0.5187	0.3011	0.2918
0.2	0.2852	0.2691	0.1802	0.1697
0.3	0.1215	0.1157	0.1358	0.1283
0.4	0.0519	0.0498	0.0512	0.0486
0.5	0.0202	0.0195	0.0205	0.0197

The issue which is studying was solved numerically using the finite difference method. Some results are shown in Figures 2-6 for different values of the parameters. The following values of the parameters were used [28-30]:

$c_0 = 0{,}01$, m^3/m^3; $v = 1 \cdot 10^{-5}$, m/s^{α}; $D_m = 5 \cdot 10^{-6}$, $m^{1+\delta}/s^{\gamma}$; $D_f = 2 \cdot 10^{-5}$, m^{β}/s^{α}; $\rho = 2500,$ kg/m^3; $\alpha_f = 2 \cdot 10^{-2}$, $1/s^{\alpha}$ $m_0 = 0{,}35$; $\alpha_m = 2 \cdot 10^{-2}$, $1/s^{\alpha}$; $T = 3600\,, s$.

In the Figures 2-6 shown profiles of c_m and s_m at the fixed points of x-axis ($x = 0.1\ m$; $x = 0.3\ m$; $x = 0.5\ m$; $x = 0.7\ m$) for different values of the adsorption coefficient.

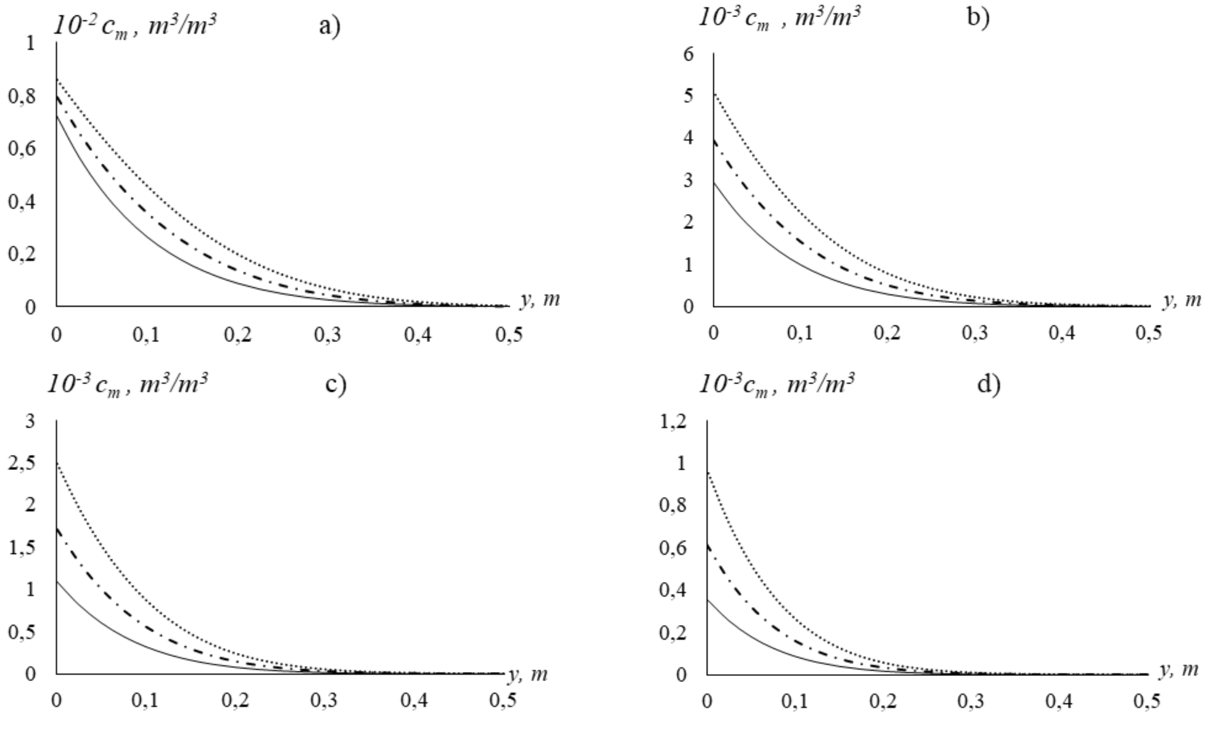
Figure 2 shows the results for the classical transport process with different values of the adsorption coefficient. According to these results, a decrease in the value of the adsorption coefficient leads to an increase in the concentration of the solute distribution (Figure 2.a-d). It was also observed that the concentration of the adsorbed substance decreases when the adsorption coefficient is reduced (Figure 2.e-h).

Figure 3 shows the results with fractional derivatives with respect to time ($\alpha = \gamma = 0.8$) in the transport equations for both zones. The results show that decreasing the order of the fractional derivative with respect to time in the transport equation below 1 slows down the diffusion of solute.

The concentration reaches $0.2\ m$ along the y-axis at $t = 3600\ s$. At large values of the adsorption coefficient, the distribution of the solute is delayed further, and the solute distributes slowly. In this case, from the values of c_m at point $y = 0$ in Figures 3. a-d, we can see that the diffusion of solute in the fracture also slowed down. 3. Figures e-h represent the concentration of adsorbed substance. Here the opposite situation is observed, that is, we can see that large values of the adsorption coefficient correspond to large values of the adsorbed substance concentration.

Figure 4 shows the results for the derivative with fractional orders with respect to the coordinate in the transport equations ($\beta = 1.7,\ \delta = 0.8$) and the derivative with respect to time with order 1. Due to the fact that the derivative with respect to the coordinate is of fractional order, we can see that a fast diffusion phenomenon occurred in both zones. However, it is observed that the concentration spread is slowed down due to the adsorption of a part of the substance entering the media.

Figures 5 and 6 consider the case where the time and coordinate derivative orders in the transport equation are combined. In Figure 5, the $\beta(<2)$, $\gamma(<1)$ cases are analyzed, and in Figure 6, the $\alpha(<1)$, $\delta(<1)$ cases are analyzed.



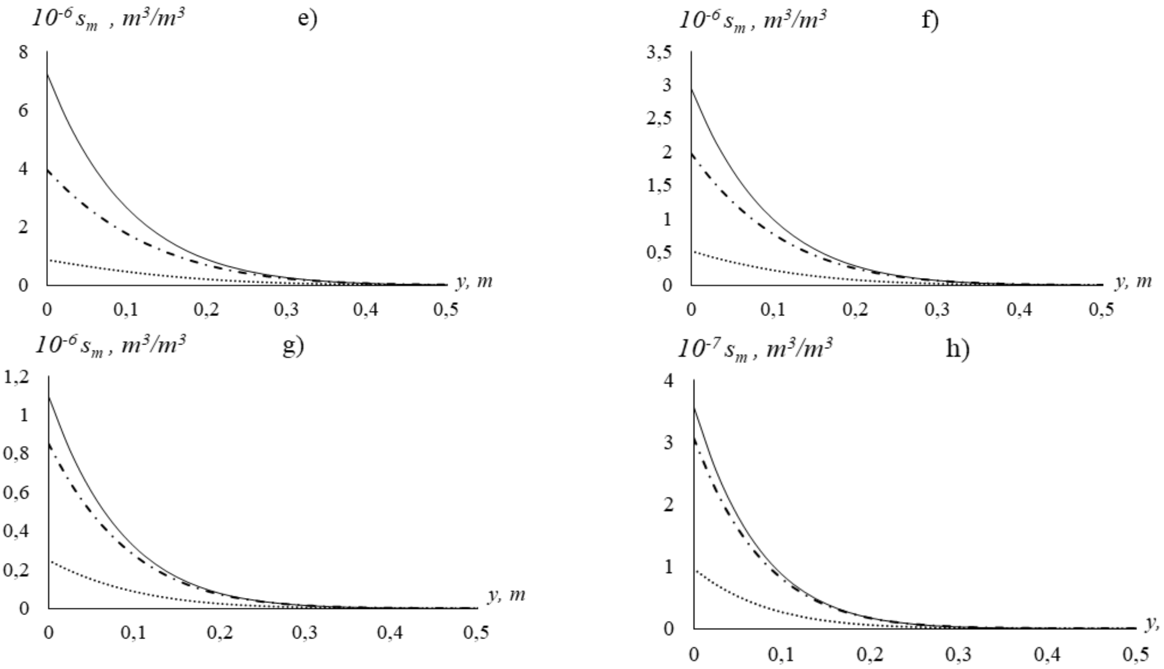
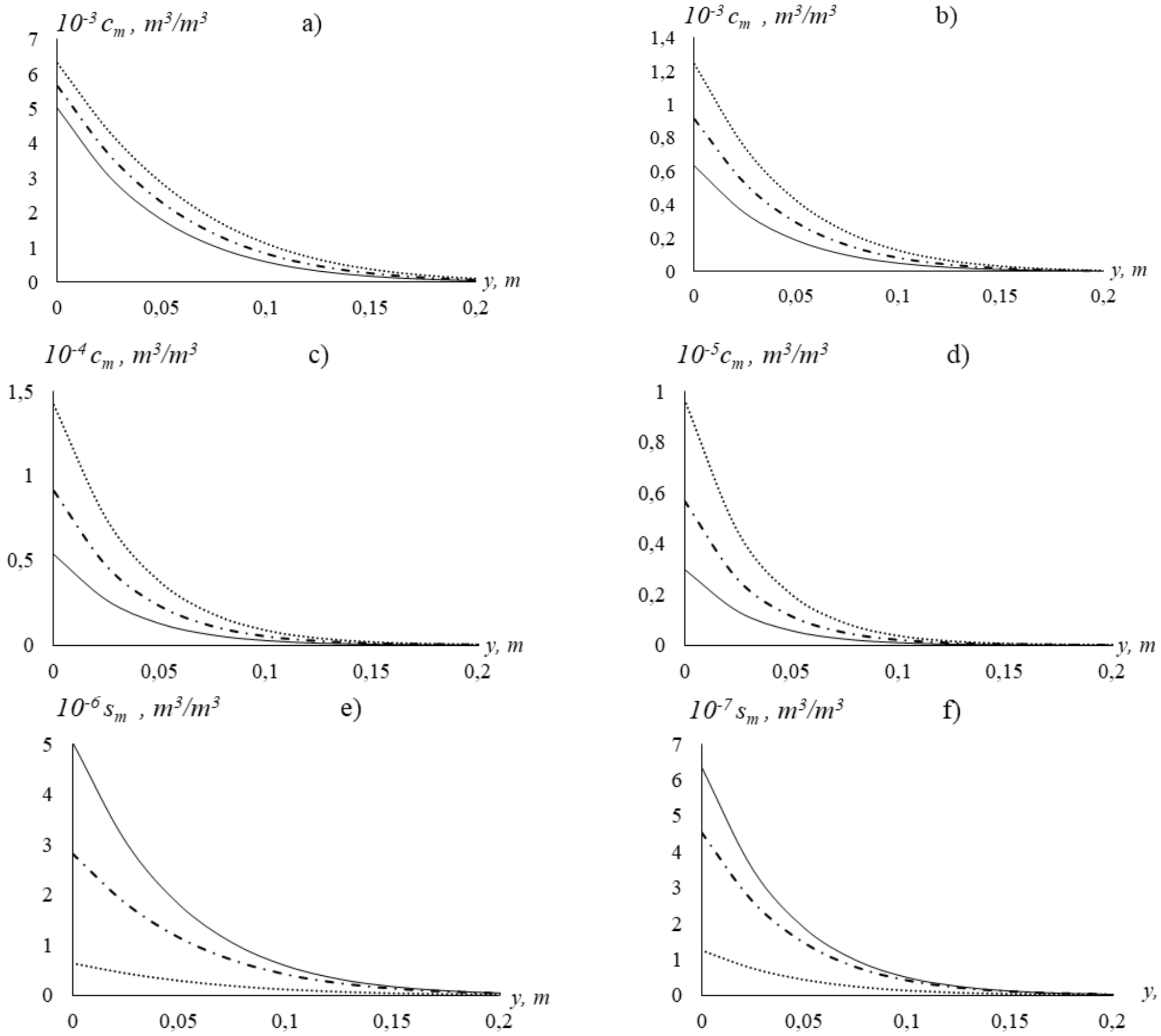


Figure 2: Surfaces sections of c_m *(a-d) and* s_m *(e-h) at* $t = 3600$ s, $\alpha = 0{,}8$, $\beta = 2$, $\gamma = 1$, $\delta = 1$, $x = 0{,}1\,\mathrm{m}$ *(a, e),* $x = 0{,}3\,\mathrm{m}$ *(b, f),* $x = 0{,}5\,\mathrm{m}$ *(c, g),* $x = 0{,}7\,\mathrm{m}$ *(d, h);* ⋯⋯ $k = 10^{-3}$, −·−·− $k = 5 \cdot 10^{-4}$, ——— $k = 10^{-4}$.



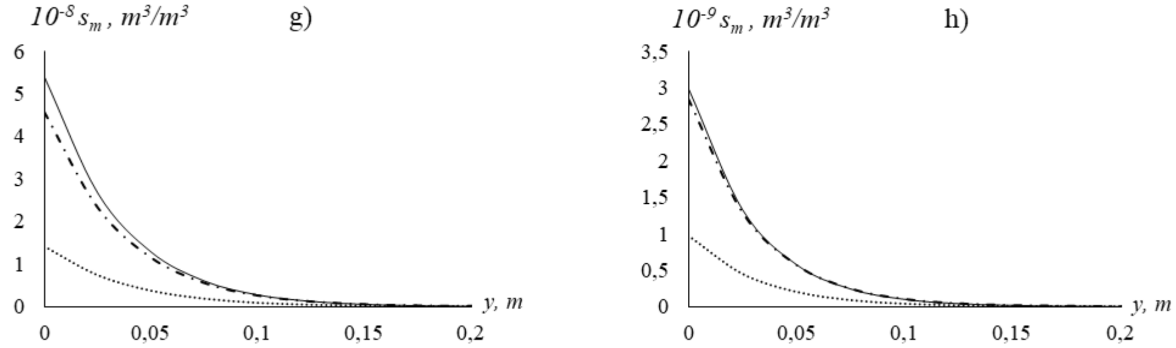
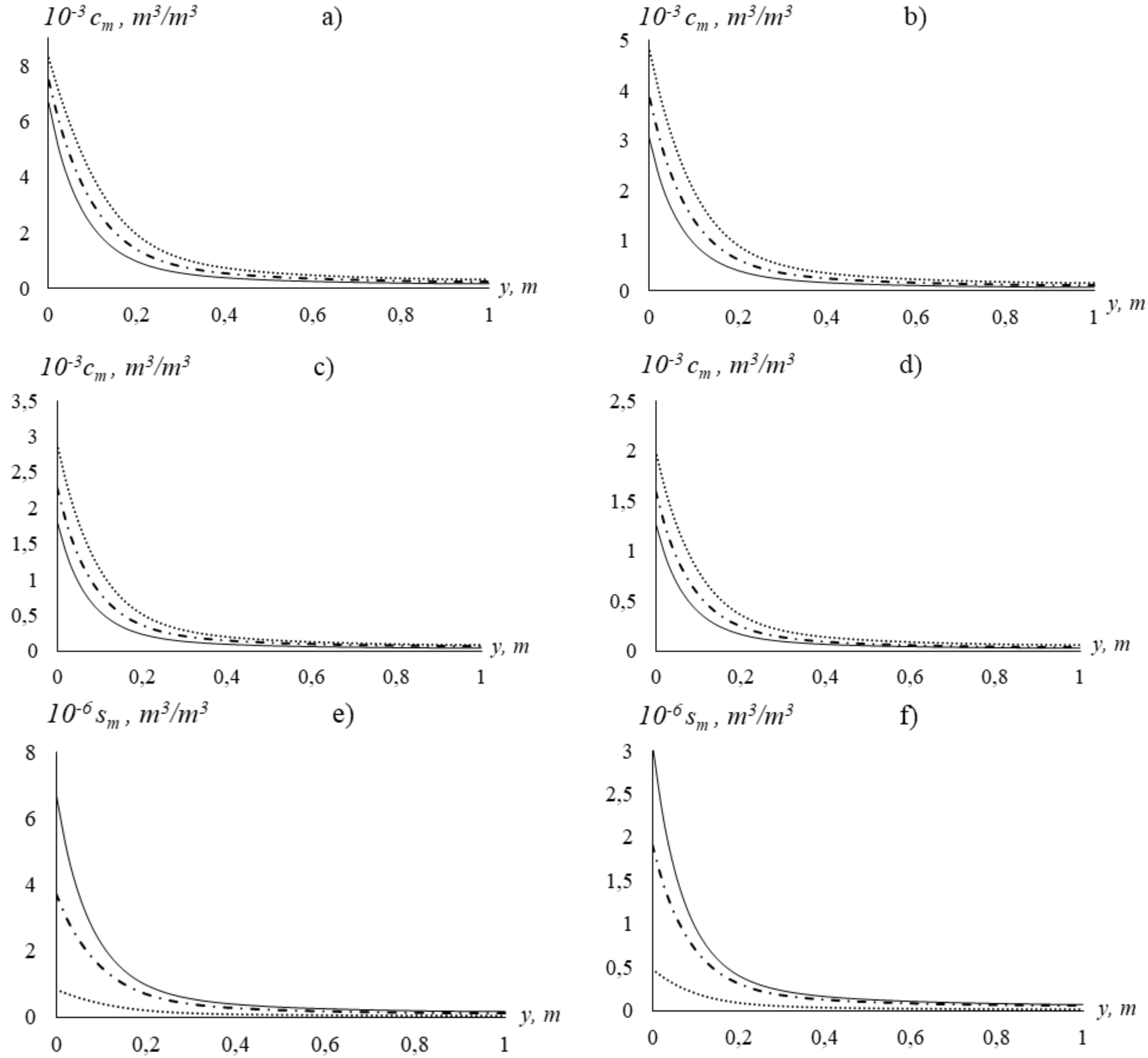


Figure 3: Surfaces sections of c_m *(a-d) and* s_m *(e-h) at* $t = 3600$ s, $\alpha = 0{,}8$, $\beta = 2$, $\gamma = 0{,}8$, $\delta = 1$, $x = 0{,}1$ m *(a, e),* $x = 0{,}3$ m *(b, f),* $x = 0{,}5$ m *(c, g),* $x = 0{,}7$ m *(d, h);* $\cdots\cdots$ $k = 10^{-3}$, $-\cdot-\cdot-$ $k = 5\cdot 10^{-4}$, —— $k = 10^{-4}$.



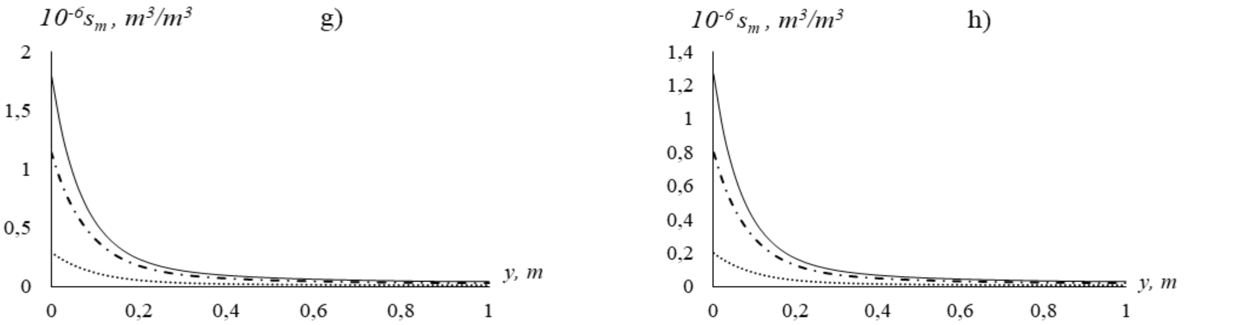


Figure 4: Surfaces sections of c_m *(a-d) and* s_m *(e-h) at* $t = 3600$ s, $\alpha = 1$, $\beta = 1{,}7$, $\gamma = 1$, $\delta = 0{,}8$ $x = 0{,}1$ m *(a, e),* $x = 0{,}3$ m *(b, f),* $x = 0{,}5$ m *(c, g),* $x = 0{,}7$ m *(d, h);* $\cdots\cdots k = 10^{-3}$, $-\cdot-\cdot- k = 5 \cdot 10^{-4}$, ——— $k = 10^{-4}$.

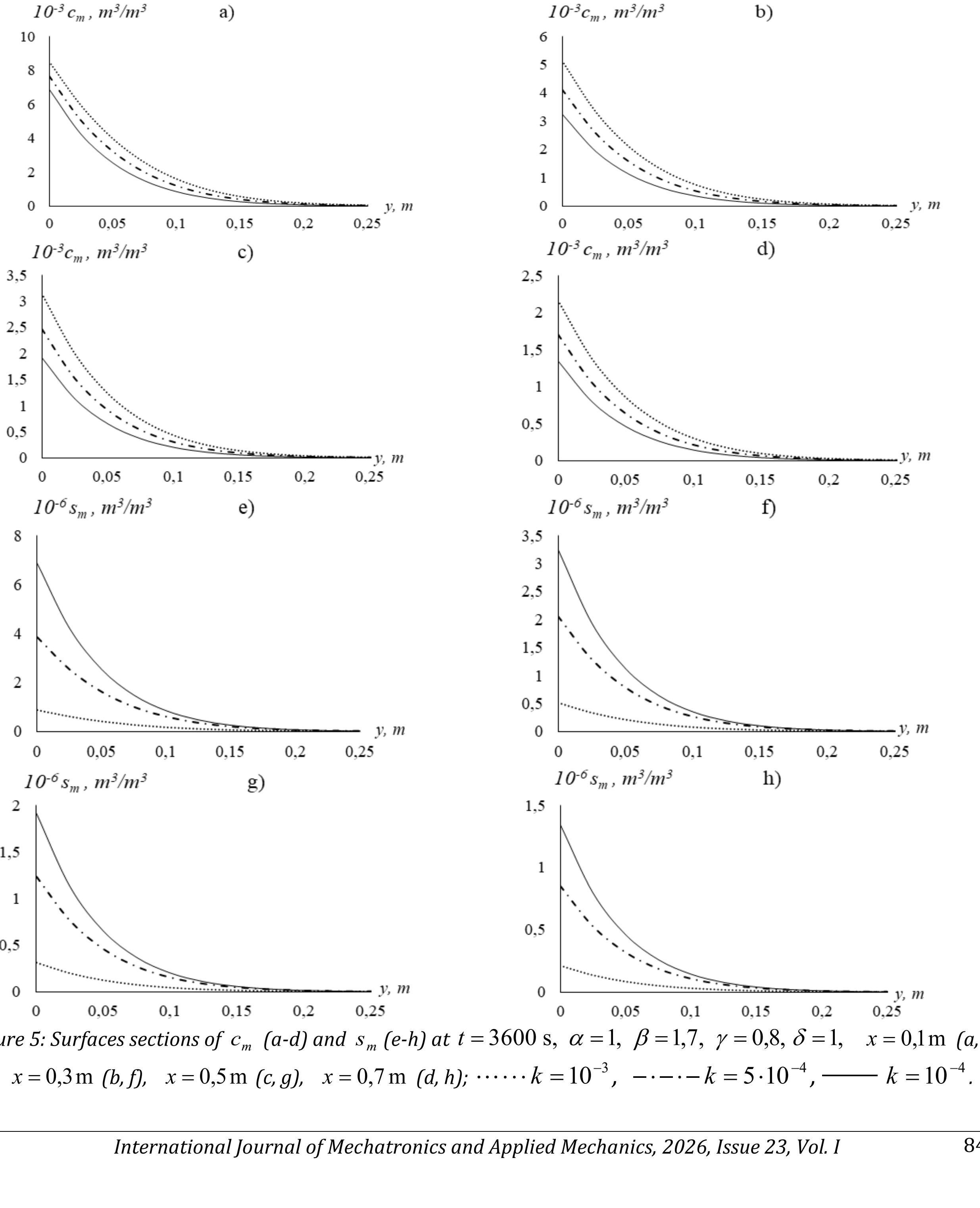


Figure 5: Surfaces sections of c_m *(a-d) and* s_m *(e-h) at* $t = 3600$ s, $\alpha = 1$, $\beta = 1{,}7$, $\gamma = 0{,}8$, $\delta = 1$, $x = 0{,}1$ m *(a, e),* $x = 0{,}3$ m *(b, f),* $x = 0{,}5$ m *(c, g),* $x = 0{,}7$ m *(d, h);* $\cdots\cdots k = 10^{-3}$, $-\cdot-\cdot- k = 5 \cdot 10^{-4}$, ——— $k = 10^{-4}$.

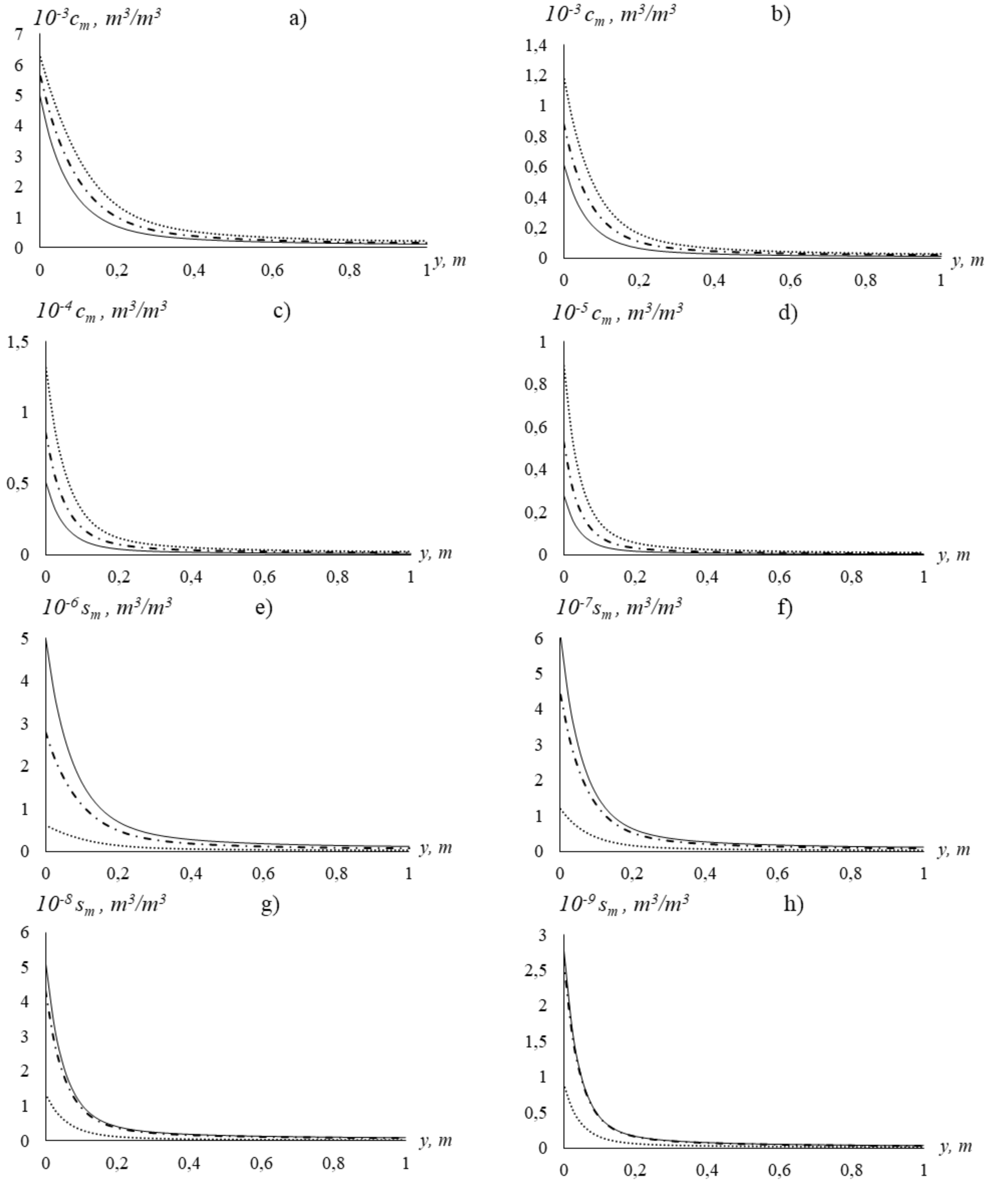


Figure 6: Surfaces sections of c_m *(a-d) and* s_m *(e-h) at* $t = 3600$ s, $\alpha = 0{,}8$, $\beta = 2$, $\gamma = 1$, $\delta = 0{,}8$, $x = 0{,}1$ m *(a, e),* $x = 0{,}3$ m *(b, f),* $x = 0{,}5$ m *(c, g),* $x = 0{,}7$ m *(d, h);* $\cdots\cdots k = 10^{-3}$, $-\cdot-\cdot- k = 5\cdot 10^{-4}$, ——— $k = 10^{-4}$.

5. Conclusions

In this work, the solute transport problem under the influence of anomaly of solute transport and non-equilibrium adsorption in the element of FPM is investigated. The medium consists of two zones: a fracture and an adjacent porous block. Solute transport takes place mainly through fractures using convective-diffusion processes, while in a porous medium only diffusion and non-equilibrium adsorption are taken into account. In the study, modeling was carried out using fractional derivatives in the sense of Caputo. This approach is especially useful in cases where traditional differential equations cannot fully describe the process.

The problem was solved using the method of finite differences. Through computational experiments, the concentration of the solute and the distribution of the adsorbed substance were analyzed at different values of the adsorption coefficient. It was observed that when the adsorption coefficient increased, the adsorption process intensified and the distribution of the solute slowed down.

The results of the study show that the use of fractional time derivatives with order smaller than 1 leads to a slower distribution of the solute. At the same time, it was found that when the order of the derivative with respect to the coordinate is reduced, the solute concentration spreads more widely, which weakens the adsorption process.

In general, this work proves the effectiveness of fractional differential equations in the in-depth analysis of anomalous solute transport in fractured-porous environments and can be widely used in applied geophysics, hydrogeology, and modeling the dispersion of pollutants.

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