

## The River System Pollutant Migration in the Context of the Sudden One-Time Discharge with Consideration of the Bottom Sediments Influence (Case of Benzene Migration in the Stryi River, Ukraine)

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### ABSTRACT

Basing on the mathematical model developed with the account of influence of bottom sediments, the parameters of benzene migration in the river caused by one-time discharge into the Stryi River were investigated. The mathematical model of migration consists of two equations that describe the movement of pollutants in the river system, taking into account the flow rate, diffusion, sorption and desorption of the pollutant by the bottom sediments of the river. The parameters of benzene distribution in the "water-bottom sediments" system were experimentally determined under laboratory conditions. With the help of computer modeling, the temporal and spatial distributions of benzene in water and bottom sediments were obtained. The regularities of benzene concentration change depending on the composition of the bottom sediments of the river have been established. The dependencies can be extrapolated to other river systems and pollutants.

**Keywords:** diffusion, mathematical model, benzene, migration, environmental safety.

### INTRODUCTION

River systems are very important and even critical elements of ecosystems in general and human life in particular. The river systems' ecological safety is largely determined by the efficiency of wastewater treatment technologies [Malovanyy et al., 2016; Shmandiy et al., 2017] and sediment utilization [Tymchuk et al., 2020], adsorption technologies for industrial wastewater treatment [Kostenko et al., 2017; Malovanyy et al., 2019] and other anthropogenic influence formation aspects. Catastrophic spills of oil and petroleum products have very significant consequences for ecosystems. [Shevchenko et al., 2021; Loboichenko et al., 2021a; Yang et al., 2021]. Restoration of the natural environment without human

intervention takes a very long time [Karabyn et al., 2019; Strelets et al., 2022]. The dangers of surface hydrocarbons are increasing with the development of new mining technologies such as shale oil and shale gas production [Lazaruk et al., 2020].

Particularly dangerous spills of hydrocarbon compounds are created in rivers near (above) drinking water intakes. Then several settlements may be deprived of drinking water supply at the same time. Therefore, the river systems water quality constant monitoring is a necessary condition [Odnorih et al., 2020; Loboichenko et al., 2021b]. Mathematical models of hydrocarbons migration in the river are an important tool to prevent such emergencies. The main requirements for such a mathematical model are its adequacy and efficiency.

Nowadays, due to the rapid development of computer technology, more and more attention is paid to mathematical modeling of various natural and man-made processes, including the dynamics of river pollution due to accidental inflow of pollutants from man-made sources [Long et al., 2016] or due to catastrophic natural processes [Whitehead et al., 2019].

One of the first models of mass transfer of pollutants in the water stream was a one-dimensional Frolov-Rodziller model, which was widely used in calculating the limits of enterprises to discharge wastewater into the river, the dilution rate of these wastewater before discharge and to solve other similar problems [Rodziller, 1984]. This model describes the change in the concentration of pollutants in the most polluted average river flow from the place of wastewater discharge to the place of complete mixing of these waters with the river. Mathematically, it is based on solving a first-order differential equation. For calculations according to this model, the final average concentration of pollutant in the area of complete mixing, the concentration of the substance in wastewater, the average flow (discharge) of these waters, the concentration of matter and water flow in the river above the wastewater discharge line, the distance from the wastewater discharge to the river station lengthwise the navigational channel, etc. are used. In addition to the mentioned parameters, which can be found in the technological and regulatory documentation, in this model the hydraulic mixing coefficient  $\beta$  should be known. The value of  $\beta$  is determined on the basis of empirical studies by formulas for different types of rivers: for plain rivers - M.V. Potapova; for all flowing reservoirs - V.M. Makaveeva. It is also necessary that the coefficient of turbulent diffusion (according to A.V. Karashev), the coefficient of friction resistance along the length (according to A. Shezi), the coefficient of roughness of the river bed (according to M.F. Silver), etc are included into the calculations [Sharp, 1981]. All these coefficients are quite generalized, so they are to be determined for each individual case.

Among other already existing mathematical models, the chamber model of mass transfer, solving diffusion problems by the Monte Carlo method and differential equations of pollution distribution are already known [Kuchment et al., 1983].

The method of L.L. Paalei and V.A. Suurkaska is based on the equation of turbulent diffusion, which is solved analytically. The formulas

for calculating the maximum concentration of non-conservative pollutants (in the general case) in any given water station, if the discharge of wastewater is at a distance  $b$  from the shore, were determined [Jorgensen et al., 2001].

The convective diffusion model is based on the solution of the one-dimensional equation of conservation of mass of solute or suspended matter.

The task is solved under the following assumptions:

- the substance is evenly distributed across the cross section of the flow, the inflow immediately spreads over the cross-sectional area;
- the substance is conservative or subject to the law of first-order reactions;
- the linear law of diffusion is valid.

The problem with the mathematical models described above is that they do not take into account the effect of bottom sediments on the mass transfer of pollutants in water. This effect will be especially significant in mountain river waters, which are characterized by small cross-sectional sizes, significant velocities and turbulence of water movement, and thus greater contact of river water with bottom sediments than in large plains. It is believed that the impact of bottom sediments on the mass transfer of the pollutant is significant, and therefore errors in estimates of the rate and concentration of mass transfer of substances in mountain river water without taking into account the influence of bottom sediments can be significant.

The purpose of the article is to study the pollutant (benzene) migration in the river system, taking into consideration the bottom sediments influence. The Stryi mountain river (Ukraine) was taken as an object of the research. It is characterized by small cross-sectional dimensions, significant velocities and turbulence of water movement, and therefore greater contact of river water with bottom sediments than in the large plain river conditions. The benzene was chosen for modeling because this chemical compound is the dominant representative of aromatic hydrocarbons, the average content of which in the oils of the Carpathian oil and gas province is 18–25%, in petrol to 7.5%, in diesel to 77% [Sirenko et al., 2017]. In addition, benzene is characterized by high carcinogenicity [Loomis et al., 2017] and resistance to biodegradation under natural conditions [Rusyn et al., 2017; Nedoroda et al., 2017]. These facts determine the high potential impact of benzene on the formation of significant risks to the stability of ecosystems, environmental and civil security.

Solving the scientific problem of developing a mathematical model for benzene pollutants distribution in a mountain river involves the formulation of the initial conditions of the model, mathematical representation, visualization of the results of its solution.

## MATERIALS AND METHODS

During the research, general scientific methods of analysis, synthesis, comparison, mathematical modeling, generalization, forecasting were used. The system of equations of the mathematical model is solved by the numerical method.

Mathematical modeling of mass transfer in a mountain river was carried out on the case of the river Stryi – the right tributary of the Dniester River (Fig. 1).

The Stryi is a typical mountain river. At the same time, the basin of the Stryi River is saturated with oil and gas wells and other oil facilities, which poses a significant threat of hydrocarbon pollution.

Samples from the middle and estuarine sections of the river were taken for the initial data of

mathematical modeling. The diffusion coefficient was calculated based on the results of particle size analysis, determination of density and other parameters of river bottom sediments.

Benzene content was determined according to the method of measuring of the mass concentration of petroleum products, in our case benzene, in surface, underground and return waters. It is based on the extraction of organic substances from a water sample with chloroform, evaporation of chloroform, dissolution of the residue in hexane, separation of polar compounds, vegetable and animal fats, light hydrocarbons on a column with aluminum oxide, hexane evaporation and gravimetric measurement of the mass of the residue. The mass concentration of petroleum products in the initial water sample is determined by the calculation method. Before measuring the mass concentration of benzene, pre-mixing was carried out with rock samples from the first supraflood terrace in a ratio (by mass) of 1:12, followed by mixing. The next stage was settling to saturate the rock with benzene and separate the liquid phase. The error of the measurement results does not exceed 10% ( $n = 3$ ).

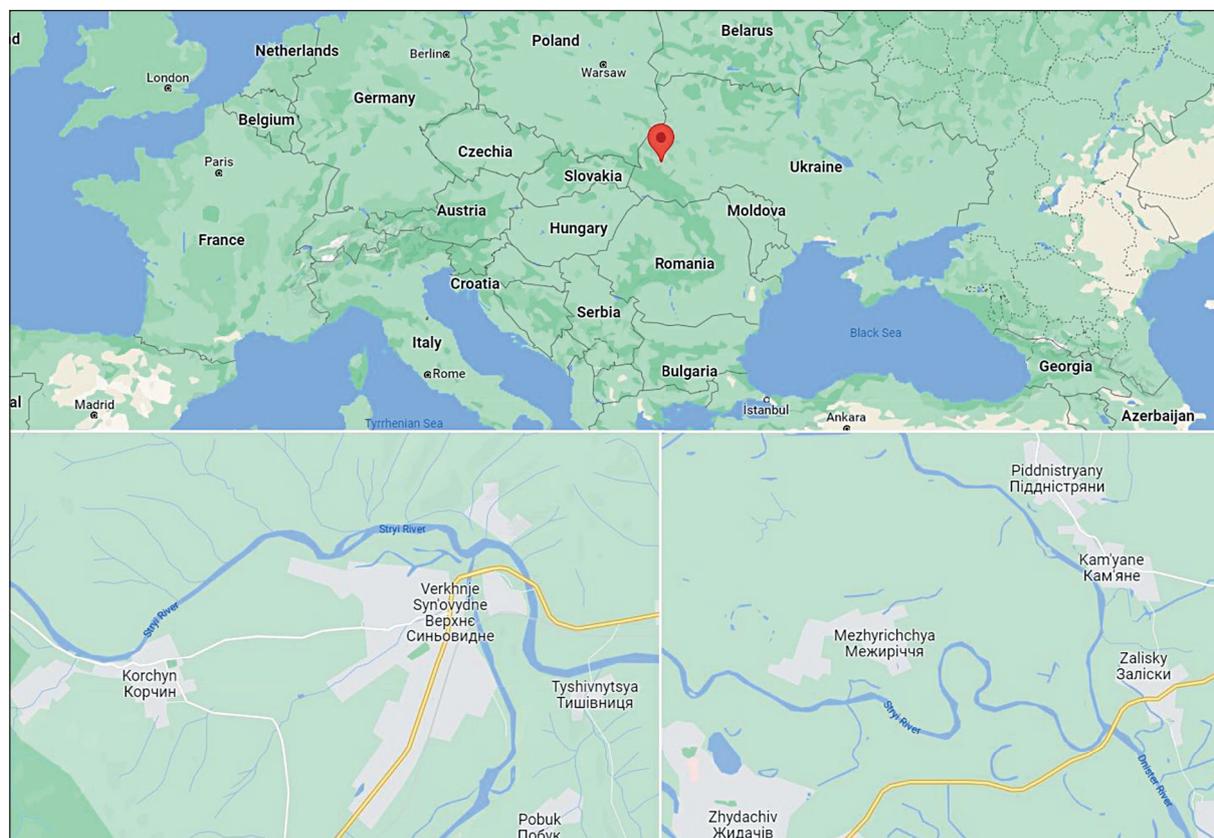


Figure 1. Sampling area

To determine the distribution coefficients of benzene in the “water-rock” (1) and “rock-water” (2) systems of the genetic horizons of the alluvial deposits of the first floodplain terrace of the Stryi River, the following formulas were used:

$$K_{w-r} = \frac{(V_w + V_b) \cdot (m_b - m_{b.w.})}{m_{s.ph.} \cdot m_{b.w.}} \quad (1)$$

$$K_{r-w} = \frac{1}{K_{w-r}} \quad (2)$$

where:  $K_{w-r}$  – the distribution coefficient of benzene in the “water-rock” system;  
 $K_{r-w}$  – distribution coefficient in the “rock-water” system;  
 $V_w$  – volume of poured water;  
 $V_b$  – volume of poured benzene;  
 $m_{s.ph.}$  – mass of the solid phase;  
 $m_b$  – mass of injected benzene;  
 $m_{b.w.}$  – the mass fraction of benzene in water after mixing.

Laboratory studies were carried out in the research laboratory of environmental safety of the Lviv State University of Life Safety (Certificate No. RL 091/21 issued November 30, 2021).

## RESULTS AND DISCUSSION

### Description of the developed model

Assume that the change in the concentration of the hydrocarbon pollutant in the river ( $C$ ) due to its one-time discharge will be influenced by diffusion, physical transfer of the pollutant due to water movement in the river and parameters of pollutant distribution in the system “water-bottom sediments”.

Then the change in the concentration of the pollutant in the river at a certain point in time ( $t$ ) can be described by the formula:

$$\frac{\partial C}{\partial t} = D_w \frac{\partial^2 C}{\partial x^2} - v_w \frac{\partial C}{\partial x} - k_r C + k_w c \quad (3)$$

where:  $C = C(x, t)$  – the concentration of the pollutant in the river water, mg/dm<sup>3</sup>;  
 $c = c(x, t)$  – the concentration of pollutants in the bottom sediments of the river;  
 $x = 0$  – reference point at time,  $t = 0$ , s;  
 $x(t)$  – the distance from the starting point, m;  
 $D_w$  – diffusion coefficient of the pollutant in water, m<sup>2</sup>/s;

$k_w$  – the coefficient of distribution of the pollutant in the “water-bottom sediments” system;

$k_r$  – the coefficient of distribution of the pollutant in the “bottom sediment-water” system;

$v_w$  – water velocity in the river, m/s.

The change in the concentration of the pollutant in the bottom sediments of the river at a certain point in time ( $t$ ) is described by the formula:

$$\frac{\partial c}{\partial t} = D_r \frac{\partial^2 c}{\partial x^2} + k_r C - k_w c \quad (4)$$

where:  $D_r$  – the diffusion coefficient of the pollutant in the bottom sediments.

Combining equations 3 and 4 we obtain the mathematical model of pollutant migration in the river system taking into account the influence of bottom sediments in the form of a system of differential equations:

$$\begin{cases} \frac{\partial C}{\partial t} = D_w \frac{\partial^2 C}{\partial x^2} - v_w \frac{\partial C}{\partial x} - k_r C + k_w c \\ \frac{\partial c}{\partial t} = D_r \frac{\partial^2 c}{\partial x^2} + k_r C - k_w c \end{cases} \quad (5)$$

### Initial conditions

The initial condition of the first equation is a function that describes a one-time discharge of the pollutant at a distance of 10 cm from the beginning of the reference. It is described by the formula:

$$C(x, 0) = \begin{cases} 1 - e^{-x}; 0 \leq x < 0,1 \\ 0; x \geq 0,1 \end{cases} \quad (6)$$

Since there is no contaminant in the bottom sediments at the initial moment of time, the initial condition for the second equation will be as follows:

$$c(x, 0) = 0 \quad (7)$$

The water velocity in the river is constant:

$$v_w = const \quad (8)$$

The chemical composition of water in the river at the site of physical modeling is stable. Lithological and chemical composition of bottom sediments in the area of physical modeling are stable.

## Boundary conditions

At the beginning of the reference we set the boundary conditions of the first kind for the concentration of the pollutant in water (9) and bottom sediments (10):

$$C(0, t) = 0 \quad (9)$$

$$c(0, t) = 0 \quad (10)$$

## Physical content of the model

The first equation describes the processes of diffusion of the pollutant in water, sorption and desorption in the “water - bottom sediments” system, taking into account the speed of the river ( $v_w$ ). The second equation describes the processes of diffusion of the pollutant in the bottom sediments and sorption – desorption in the “bottom sediments – water” system.

## Model input

The diffusion process in binary solutions is characterized by the mutual diffusion coefficient ( $D_{AB}$ ), which has a dimension of  $m^2/s$  and depends on the temperature, size and shape of diffusing molecules, the ability to associate and dissociate and solvent parameters such as viscosity, properties of diffusion medium molecules to the association [Sherwood et al., 1975]. Based on the model according to which the particle of solute is considered as a sphere moving through a continuous medium of solvent, the Stokes-Einstein equation was obtained:

$$D_{AB} = \frac{kT}{\sigma\pi r_A \eta_B} \quad (11)$$

As well as Auerbach’s empirical equation:

$$D_{AB} = KM_A^{-b} \quad (12)$$

where:  $k$  – the Boltzmann constant;  
 $r_A$  – the radius of the diffusing spherical molecule;  
 $\eta_B$  – the viscosity of the solvent;  
 $M_A$  – the molar mass of the diffusing molecules;  
 $K, b$  – the empirical constants.

The constants  $K, b$  in equation (12) depend on the chemical nature of the diffuser [Rudobashta et al., 1993].

For practical calculations, the diffusion coefficient in mixtures of non-electrolyte liquids uses the Wilkie-Cheng formula [Rudobashta et al., 1993; Sherwood et al., 1975]:

$$D_{AB}^0 = 7,4 \cdot 10^{-8} \left[ (\varphi M_B)^{1/2} \frac{T}{\eta_B V_A^{0.6}} \right] \quad (13)$$

where:  $D_{AB}^0$  – the coefficient of mutual diffusion of solute  $A$  at its very low concentrations in solvent  $B$ ,  $cm^2/s$ ;  
 $M_B$  – molar mass of substance  $B$ ;  
 $T$  – temperature,  $K$ ;  
 $\eta_B$  – viscosity of substance  $B$ ,  $mPa \cdot s$ ;  
 $V_A$  – molar volume of solute at normal boiling point,  $cm^3/mol$ ;  
 $\varphi$  – the association parameter of solvent  $B$  (which has a value of 2.6 for water).

Accordingly, coefficient of benzene ( $C_6H_6$ ) in water is  $1.02 \cdot 10^{-9}$ ,  $m^2/s$ , in water-saturated pebble-sand mixture (middle part of the river Stryi)  $5 \cdot 10^{-10}$   $m^2/s$  and in water-saturated sand-clay mixture (estuary of the river Stryi)  $4 \cdot 10^{-10}$   $m^2/s$ .

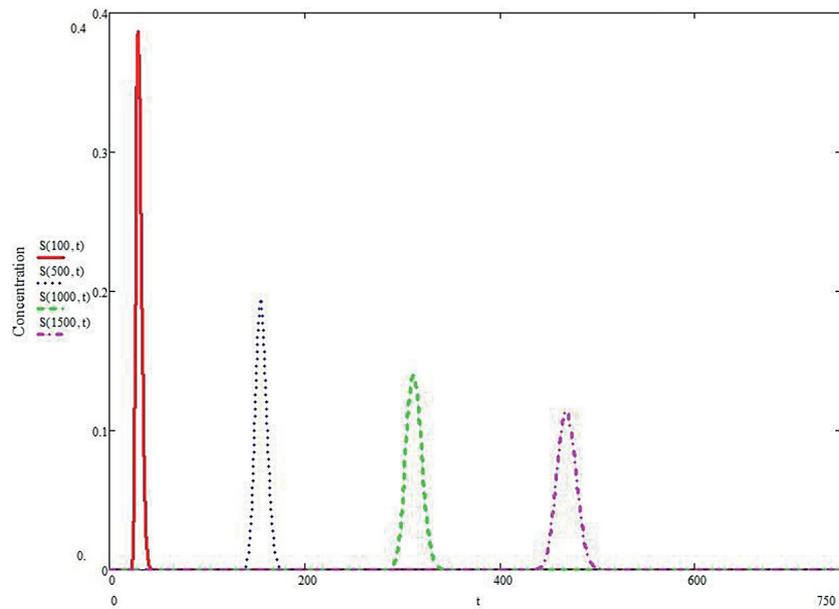
The concentration of benzene at the initial time, for the convenience of further use of the results of mathematical modeling is reduced to  $1 \text{ g/dm}^3$ .

To quantify the adsorption of bottom sediments, the partition coefficient  $k_r$  in the bottom sediment-water system is used, which represents the ratio of the concentration of the active substance adsorbed by the bottom sediments  $c$  ( $mg/dm^3$ ) to its concentration in water  $C$  ( $mg/dm^3$ ) at equilibrium  $k_r = c/C$  in the case where the concentration of the active substance in both phases is expressed in units of weight (weight/weight)  $k_r$  is a dimensionless quantity. Accordingly,  $k_w$  is the distribution coefficient in the “water-bottom sediments” system. The coefficients  $k_r$  and  $k_w$  are dimensionless quantities.

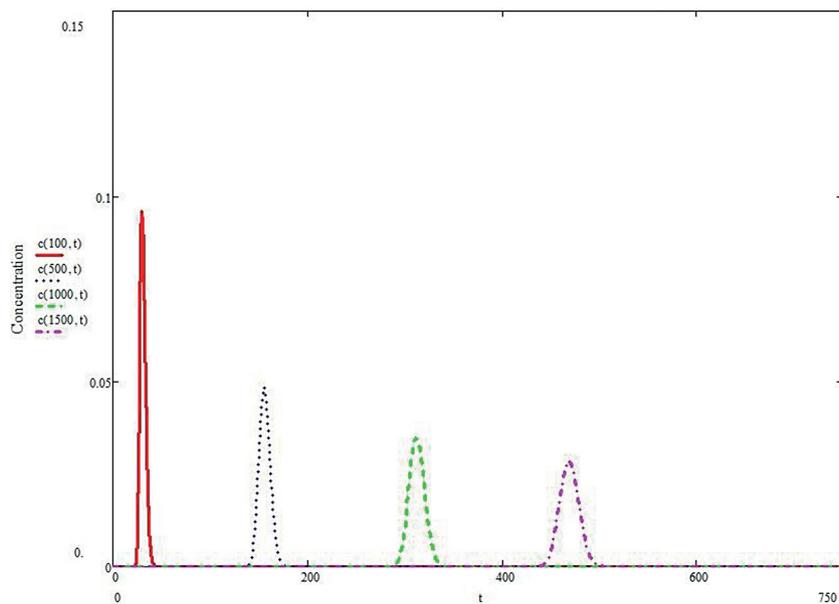
The partition coefficients of benzene in the system of water-bottom sediments were calculated basing on experimental data in the amount of 0.50:2.00 in middle part of river Stryi and 0.75:1.33 in estuary of the river Stryi.

## Numerical solution of a mathematical model

Numerical solution of the system of equations for the given values of diffusion coefficients, flow rate, sorption and desorption is performed for benzene and is presented in graphs of the concentration of the pollutant over time at a certain



**Figure 2.** Changes in the concentration of benzene over time (t) in the water of the middle part of the River Stryi



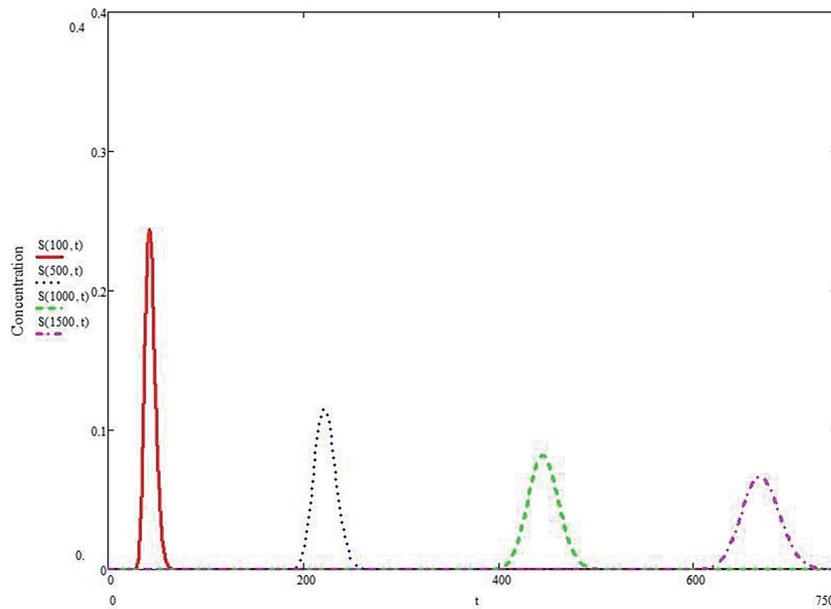
**Figure 3.** Changes in the concentration of benzene in the bottom sediments over time (t) in the water of the middle part of the River Stryi

distance from the source of pollution in water and bottom sediments. Changes in the concentration of benzene in water and bottom sediments over time are shown in Fig. 2–3.

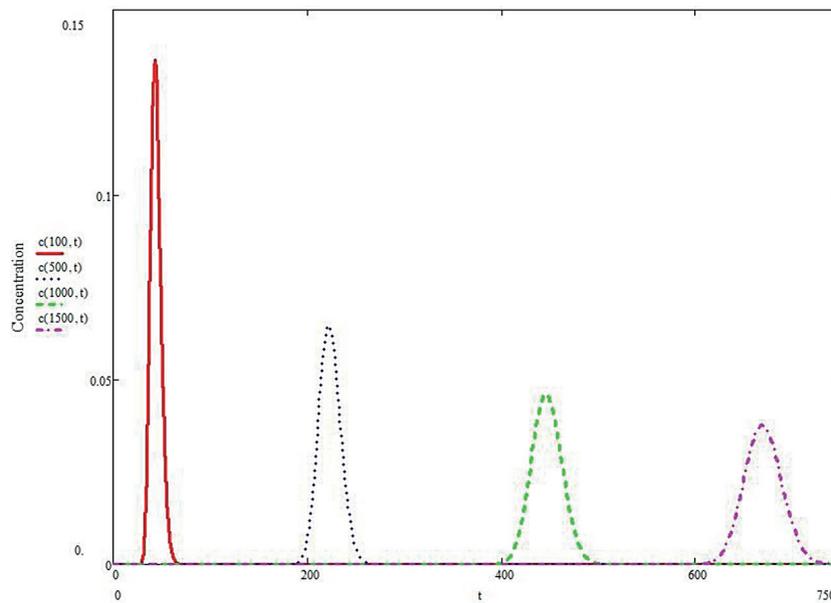
Analyzing the data presented in the graphs, it should be noted that at a distance of 100 m from the source of pollution graphs of benzene concentrations increase rapidly in water to  $0.39 \text{ g/dm}^3$  after 30 s from the discharge of the pollutant, and then fall to zero after 41 s. From a distance from the source of pollution, the graphs of changes in benzene concentration over time reduce their

peaks, become flatter and more symmetrical. At a distance of 500 m from the source of contamination, the maximum benzene content is reached at 155 s after discharge of the contaminant and is  $0.19 \text{ g/dm}^3$ . At a distance of 1500 m from the source of pollution, the maximum content of benzene is reached as much as 466 s (7.8 min) after the discharge of the pollutant and is  $0.11 \text{ g/dm}^3$ .

In the bottom sediments of the middle parts of the Stryi River, the concentration of benzene is lower compared to the concentration of benzene in water (Fig. 3). This is due to the fact that much



**Figure 4.** Changes in the concentration of benzene over time (t) in the water of the estuary of the River Stryi



**Figure 5.** Changes in the concentration of benzene in the bottom sediments over time (t) in the estuary of the River Stryi

of the pollutant is washed away by the surface waters of the mountain river and benzene is lighter than water. The configuration of the graph of the change in the concentration of benzene in the water is similar to the configuration of the graph of the change in the concentration of benzene in the bottom sediments.

Similar dependences were obtained for the estuary of the Stryi River (Fig. 4–5). However, different parameters of water velocity and bottom sediment composition, which was reflected in the values of the parameters of distribution of

hydrocarbon pollutants in the water-bottom sediment system, were shown in the graphs.

The concentration of benzene in the water of the estuary of the River Stryi at a distance of 100 m from the source of discharge reaches 0.24 g/dm<sup>3</sup>, which is 0.15 g/dm<sup>3</sup> higher than this value in the middle part of the river. The water of the estuary of the River Stryi is more slowly purified from benzene compared to the intensity of purification from this contaminant in the middle part. This is probably due to the lower speed of the river.

The maximum content of benzene in the water of the mouth of the Stryi River at a distance of 1500 m from the source of pollution is reached only in 667 s (11.11 min) after the discharge of the pollutant, which is one third longer than in the middle part of the river.

The benzene concentration in the bottom sediments of the waterway of the river Stryi and in its middle part is lower than in water. At the same time, the concentrations of pollutants at the same distances from the beginning of the reference as in the middle reaches of the river are also slightly higher than in water.

Note that the developed mathematical model of benzene migration in the system of water-bottom sediments of the river makes it possible to predict the spread of this pollutant in both water and bottom sediments.

We acknowledge Dr. Leonid Sysa for his kind consultations in carrying out the laboratory investigations and discussion of its results.

## CONCLUSIONS

1. A mathematical model of distribution of pollutant hydrocarbon compounds in the river due to its one-time discharge in the system of water - bottom sediments is described by a system of two differential equations. The first equation describes the pollutant diffusion of the in water, sorption and desorption in the system of water - bottom sediments, taking into account the speed of the river ( $v_w$ ). The second equation describes the pollutant diffusion in the bottom sediments and sorption - desorption in the system of bottom sediments - water.
2. Numerical solutions of the mathematical model are obtained, which are presented in graphs of dependences of benzene concentration on time. Such dependences are obtained for two sections of the river: middle part and estuary of the River Stryi.
3. The regularities of the pollutant distribution depending on the composition of the bottom sediments of the river and its flow are substantiated.

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