Development of some Models for Assessment the Dynamic Migration Processes of Phosphates in Soil Columns

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Summary

Due to complexity of phosphate (P) migration in soil columns, the problem of theoretical approach of modeling the process is specialy difficult. So, numerous semi-empirical models based on admission of simplifying assumptions were developed, some of them being apart from the real phenomenon.

In this paper are presented the breakthrough curves, the phosphate (P) usage degree in soil, based on a dynamic study, realised in order to determine phosphate sorption rate on the granules of two soil types (cambic chernozem and psamosoil) from Romania. Due to random character of the process, which is influenced by many naturally variable factors, this can be addressed from stochastic point of view. Experimental data regarding phosphate migration in soil column evolution process are represented like curves with sigmoid profile. Based on this observation, Rosin-Rammler and Schuhman sigmoid profile curves, known models and logistic type equations were used as simulation models representing phosphate transport and sorption kinetics for laboratory experiments in columns.

Testing the models with the experimental data resulted with the conclusion that the most appropriate models describing the process of phosphate migration in soil columns are Rosin-Rammler and logistic type equation, $C = 1-\exp(-bt^n)$ and $C = [1+\exp(\alpha+\beta t)]^{-1}$, (C – relative concentration; t – time; b, n, α, β - constant coefficients determined from experimental data by non-linear regression) for which the correlation coefficient is $R^2 \geq 0.983$ for both types of soil.

Key words

sorption of phosphates, mathematical model, soil, dynamic regime

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Introduction

Sorption processes are among the most important chemical processes in soils that affect nutrient transport. Removal of phosphate (P) from soil by the plant's absorption, by leaching or by other processes related to soil dynamics, results in the reduction of P content as mobile and gradual decline of the production capacity of soils (Nohra, 2006). Sorption contains two processes: adsorption and desorption. Adsorption occurs when phosphate ions are removed from solution and attached to soil particles surface. If phosphate ions are adsorbed inside the particle and then disseminate they are absorbed In case when they run from inside of the particle to outside then they are desorbed.

Phosphate anions can adsorb on the surface of soil particle by specific adsorption reactions (nonelectrostatic forces) and/or may precipitate in soil aggregates. Sposito (1982) defines the specific adsorption of phosphate anion as complexation inside a complex ball and Tan and Dekter (1998) defines it as ligand exchange or chemosorption. Specific adsorption reaction is the formation of covalent bond complexes in monodentate between phosphate anions present in solution and metal complexes from soil while OH^- groups are replaced at soil particle surface (Sposito, 1982; Tan and Dekter, 1998).

The phosphate anion adsorbtion in the soil solution is highly nonlinear, because the energy levels vary among different centers of binding the surface soil particles. Thus, high-energy centers are occupied before the low energy ones. This non-linearity is mathematically presented by many alternative equations.

Thus, many theoretical models have been developed in order to describe the phenomenon of migration of phosphate anion in soil columns.

An analytical equation Gaussian distribution type after a single dimension (x axis at time t) for various ions transport in soil is proposed by Hunt in 1978 (Ramaswami et al., 2005):

$$C(x,t) = \frac{M_a}{2n\sqrt{\pi t D_L}} \exp\left[-\frac{(x-vt)^2}{4D_L t}\right] \quad (1)$$

where: $C(x,t)$ represents P concentration in solution on x axis, at time t; M_a = mass/area; D_L – longitudinal dispersion coefficient; t - time; n – soil porosity; v – Darcy velocity.

Chen et al. (1996) proposed two identical kinetic forms in versions of the Freundlich differential equation to present two parallel processes of slow and fast sorption for the analysis of the experiments in column in which P is sorbed and P concentrations in solution are measured at different depths after application of phosphate solution at the soil surface (Mansell et al., 1985; McGechan et al., 2002):

$$\frac{\partial Q}{\partial t} = \frac{\theta}{\rho} k_1 C^n - k_2 Q \quad (2a)$$

$$\frac{\partial S}{\partial t} = \frac{\theta}{\rho} k_3 C^n - k_4 S \quad (2b)$$

where total sorption S_{tot} is given by ecuation (3) and: $Q = f \cdot S_{tot}$; $S = (1-f) \cdot S_{tot}$, where f is a fraction of the center sorption taking part in the rapid sorption, θ – the volumetric water content of

the soil, ρ –the bulk density of the soil, C – the concentration of P in solution, k_1, k_2, k_3, k_4 – the rate of coefficients for forward and backward reaction, n – reaction order.

Enfield and Shew (1975), Enfield et al. (1976, 1981) and Stuanes and Enfield (1984) have used the equation below, with different versions of the equations, for rapid response sorption and slow reaction to represent the migration of phosphorus in soil samples:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial z} - \frac{\rho}{\theta}\frac{\partial S_{tot}}{\partial t} \quad (3)$$

where θ and ρ have the same physical significance as above, v is the interstitial water velocity, z is distance and $S_{tot} = Q + S$ and D is the solute dispersion coefficient.

The models presented above for sorption processes were developed as simulation models representing sorption kinetics and P transport. These equations are usually used for experimental results analysis obtained for the laboratory research done in soil columns.

The usage of any of the models presented has major drawbacks due to difficulties in analyzing the experiments.

In this respect, in this paper, phosphate anion migration while phosphate aqueous solutions were traveling through columns of cambic chernozem and sandy soil was experimentally studied in laboratory conditions.

This phenomenon was described by the calculated parameters and by using the Rosin-Rammler, Schuhman models and logistic type equation that has been tested with experimental data whose curves were compared with experimental breakthrough curves, identifying the most appropriate model.

Theoretical aspects

Addressing the development of theoretical models for phosphate (P) migration in soil columns involves particular difficulties due to the complexity of process and many naturally variable factors that affect it and cannot be controlled. Therefore, in addition to developing semi-empirical models based on admission of some simplifying assumptions, some of them departing significantly from the real phenomenon may develop some empirical models based on analysis, interpretation and processing of data obtained from experiments.

We will present some considerations on how to develop some empirical models related to this process. Given the random nature of the process that is influenced by many naturally variable factors and impossible to control, it can be approached from stochastic point of view.

The corresponding points of experimental data on changes in the migration process of phosphates in soil columns have the curves with a sigmoid profile (see Figure 1). Also, the process development in time is of the cumulative type, tending to a state of saturation (balance) after a certain time, the growth was faster in the first period, followed by a slow increase until the cancellation of the second period, which gives the sigmoid shape to the corresponding curve (Figure 1). This profile is similar to the one of cumulative granulometric curve on the weight percent of a solid material sample in granular condition of particles with size smaller than a required value.

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The change in the weight (expressed in decimal) is the most appropriate described by one of the most used three stochastic models known (Henderson et al., 1968; Căsăndroiu et al., 2004) namely:

– Rosin Rammler model type:

$$f(x) = 1 - \exp(-bx^n) \tag{4a}$$

– Schuhman model type:

$$f(x) = \left(\frac{x}{k}\right)^a \tag{5a}$$

– Logistic function model:

$$f(x) = [1 + \exp(\alpha + \beta x)]^{-1} \tag{6a}$$

where: $f(x)$ represents mass weight (expressed in decimal) of particle fraction with size smaller than x; $a, k, b, n, \alpha, \beta$ - coefficients determined from experimental data by non-linear regression

Given the above stated analogy to describe the process of phosphates (P) migration in soil columns, we proposeed testing the three types of stochastic models, namely:

– Rosin Rammler model type:

$$C = 1 - \exp(-bt^n) \tag{4}$$

– Schuhman model type:

$$C = \left(\frac{t}{k}\right)^a \tag{5}$$

– Logistic function model:

$$C = [1 + \exp(\alpha + \beta t)]^{-1} \tag{6}$$

where: C is relative concentration (reported to initial concentration, C_0); t – time; $b, n, k, a, \alpha, \beta$ - constant coefficients determined from experimental data by non-linear regression.

These three models were tested with experimental data in order to determine the most suitable model.

Thus, the proposed models, in particular their parameters can represent input data for the preparation of mathematical models to more fully describe the behavior of phosphate ion in soil in dynamic conditions.

Materials and methods

Experiments were conducted on two soil types, cambic chernozem and psamosol from Teleorman and Dabuleni area, Românіа. The soil from Teleorman area is a soil with a lute-clay texture, known in the literature as chernozem clay (cambic), and the soil from Dabuleni area is a sandy soil (psamosoil) with a (%) content of granulometric fractions presented in Table 1. Physico-chemical properties of the two soil types and the analytical methods used are presented in Table 2. The soil from Teleorman has a characteristic reaction area of transition from strongly acidic to moderate, while the one from Dabuleni has a neutral to weak basic reaction (Table 2).

The equipment used was composed of: Shimadzu AW 220 analytical balance having 0.0001 g precission; double beam UV-VIS spectrophotometer Cintra 5 (spectral domain, 190-1100 nm), optical cells of glass / quartz with 10 mm thickness and pH-meter Consort C830 (0.01 pH resolution, Pt 1000 sensor).

Table 1. Classification of particle size fractions (STAS 7184/10-79)

Granular fraction name	Grain diameter (mm)	Composition (%)	
		Teleorman Soil	Dabuleni Soil
Coarse sand	>0.2	0.35	66.0
Fine sand	0.2 – 0.02	17.21	21.0
Dust	0.02 – 0.002	39.52	5.6
Clay	<0.002	42.92	7.4

Table 2. The physico-chemical properties of cambic chernozem soil (Teleorman area) and sandy soil (Dabuleni area)

Property	Value		Analysis method (symbol)
	Teleorman soil	Dabuleni soil	
pH 1:2.5	5.4	7.2	SR ISO 1039:1999
Humus (%)	2.9	0.5	STAS 7184/21-82
$CaCO_3$ (%)	0.00	0.00	STAS 7184/16-80
N_{total} (%)	0.18	0.065	SR ISO 11261:2000
P_{AL} (mg kg^{-1})	90.00	8.400	STAS 7184/19-82
P_{total} (%)	0.090	0.028	STAS 7184/14-79
K_{AL} (mg kg^{-1})	315	34.00	STAS 7184/18-80
Zn (mg kg^{-1})	3.69	-	SR ISO 11047:1999
Cu (mg kg^{-1})	3.01	-	
Fe (mg kg^{-1})	40.37	2.00	
Al (mg kg^{-1})	170	2.00	
Mn (mg kg^{-1})	4.5	-	
Alkali saturation level (%)	83.2	86.8	SR ISO 11260:2001
Exchange alkali (me/100 g sol)	28.3	2.27	SR ISO 11260:2001
Hygroscopicity coefficient (%)	6.9	1.900	STAS 7184/6-87
Volumetric weight (g cm^{-3})	1.25	1.38	SR ISO 11272:2000

Characterization of these soils has been produced and made available by Agrochemical and Pedological Research Institute (ICPA), Bucharest (Romania)

Reagents used in this study were prepared according to the Romanian protocol ISO (Romanian Institute for Standardization) with symbol SR 11411-2:1998: nitric acid 63%, $\rho=1.41$ g cm^{-3}, color reagent: vanado molybdate reagent, KH_2PO_4 solutions of 60 mg L^{-1} concentrations, and for pH adjustment was used sodium hydroxide 2M.

As aqueous phase was used a monopotassium phosphate solution (KH_2PO_4) 60 mg L^{-1}, maintaining uniform flow throughout the experiments; the phosphate anion ($H_2PO_4^-$) was noted with P.

Soil columns used in miscible displacement experiments of monopotassium phosphate synthetic solution were obtained by manually adding the air dried soil. The columns used had cylindrical shape, and were made of glass with: inside diameter of 3.5 cm, total lenght of 50 cm, and the soil layer height was equal to the height of the arable layer, about 30 cm (Figure 1). After filling, the columns were saturated with KH_2PO_4 solution with concentration of 60 mg L^{-1}, and the effluent was collected in
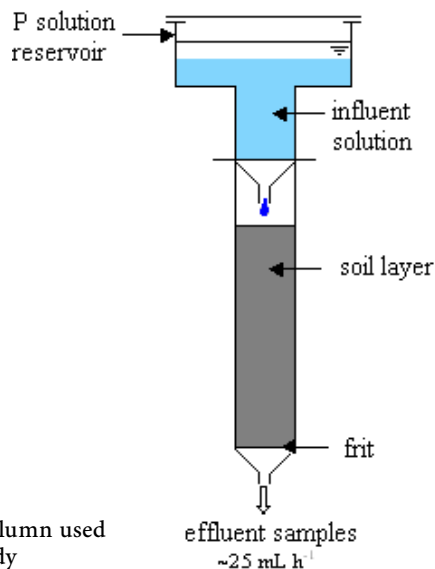
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Figure 1.
Experimental column used
for dynamic study

Table 3. Significant experimental data (extract) regarding P absolute concentration variation in time for the two types of soil, $C_0 = 60$ mg L^{-1}

Cambic chernozem soil -Teleorman		Sandy soil - Dabuleni	
Time (h)	C (mg L^{-1})	Time (h)	C (mg L^{-1})
0	0	0	0
30	0	1	2.96
34	2.38	3	4.28
51	7.25	6	6.91
55	7.95	7	8.88
97	17.45	9	12.83
101	19.4	10	15.46
105	23.35	18	27.3
123	27.3	24	40.46
128	29.95	28	47.7
144	35.5	30	49.01
145	39.14	43	51.65
150	42.43	46	52.96
171	47.04	55	53.62
190	52.96	67	56.9
213	56.91	70	58.2
218	58.88	74	59.5
220	60	75	60

25 mL fractions in continuous stream with almost 0.42 mL min^{-1} flow. Under those conditions, the soil column was continuously fed with constant concentration solution (solution ion) and reaction products were continuously collected and removed in effluent until the equalization of phosphate concentration from influent with the one in the effluent. The experiments were done in the same operational conditions and the number of experiments varied depending on the rate of P concentration increase in effluent and on the soil type. So, for cambic chernozem samples were collected during nine days (25 mL samples and average samples obtained by addition of continuous 25 mL collected samples in the moment when the concentration didn't record significant changes), and for sand soil the samples were collected during three days.

Mobile P content determination was done according to Romanian ISO protocol (SR 11411-2-98, 1998). The work was done at room temperature (20±2ºC), the solution pH was 5, it was adjusted with sodium hydroxide, and a continuous flow was mantained. The effluent fractions collected were analised by spectrophotometrical method (vanado molybdate reagent) at 470 nm wavelength.

Results and discussion

The experimental data obtained in the working conditions mentioned above are presented in form of breakthrough curves as variation of P concentration in the effluent in time for the two types of soil, in Table 3 (in extract). During the experiments, we observed that, at prolonged operation times, two areas of soil layer were formed: P ions saturation area, placed in the upper region of the column and mass transfer area placed in the lower side, with evident tendency to diminish until disappearance. In this case, the P ion concentration in the effluent starts to rise and finally reaches the initial concentration value (C_0).

Further passage of the liquid flow over the soil layer doesn't produce concentration changes neighter in liquid phase nor in solid phase, saturation balance state settlement in dynamic conditions is taking place.
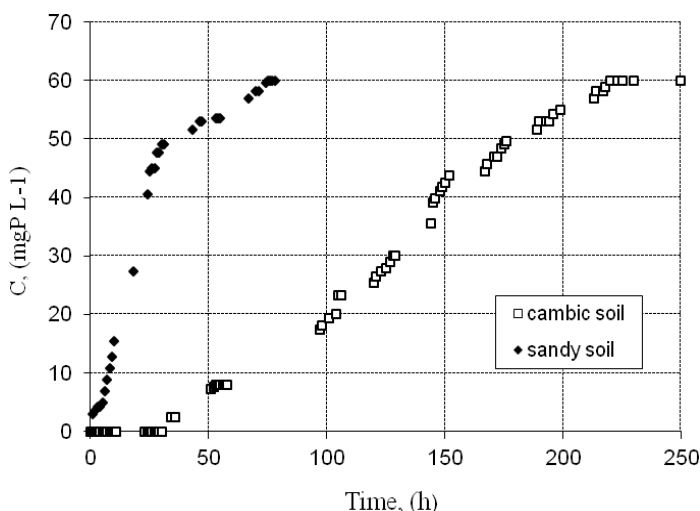

Figure 2. Effluent P concentration variation in time

Experimental data used to draw the breakthrough curve are presented in Table 3.

Experimental data from Table 3 regarding fixed bed ion exchange process for the two types of soil are graphically represented in Figure 2.

The way in which experimental data were laying in the graph for the two types of soil used in our experiments, suggests that they are placed on sigmoid type curves, for which are known corresponding models and further the suitability to use them were tested.

The data from Table 3 allow us to evaluate the characteristic parameters of kinetic study in dynamic regime, with different parameters as follows (Orbeci, 2005; Untea, 2004):

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– evolution time of mass transfer zone t_e (h):

$$t_e = t_S - t_s \qquad (7)$$

where: t_s = breakthrough time (represents the time at which the P concentration in effluent value starts to rise), (h); t_S = saturation time (represents the time at which the effluent and influent concentration are equal), (h);

– formation time of the mass transfer zone t_f , (h):

$$t_f = (1-f)t_e \qquad (8)$$

where: f = m/(m+n) is the factor defined as the ratio between retained P quantity during mass transfer zone evolution (m – the surface placed above breakthrough curve between t_s and t_S and n – the surface placed under the breakthrough curve between t_s and t_S); the value of this factor is found in the range of 0.30÷0.65 (Untea, 2004);

– mass transfer zone lenght, $L_{ZTM,}$ (cm):

$$L_{ZTM} = L \cdot \frac{t_e}{t_S - t_e(1-f)} \cdot \phi^1 \qquad (9)$$

where: L represents the lenght of soil fix bed, ϕ is a form factor;

– soil usage degree θ_u, (%):

$$\theta_u = 1 - \frac{L_{ZTM}}{L} \cdot (1-f) \qquad (10)$$

Based on the data presented in Table 3 and using the equations (7)÷(10) the characteristic parameters of ion exchange process in dynamic conditions for the two types of soil were evaluated and presented in Table 4.

From kinetic point of view and by analyzing the values presented in Tables 3 and 4 it can be observed that the necessary time to equalize the concentration from influent and effluent was high, almost nine days in case of cambic chernozem soil and three days in case of sandy soil. In other words, when the saturation state is hard to achieve, the border zone (the area situated between $t_{1/2}$ şi t_S) becomes larger and very visible in the case of cambic chernozem soil.

The structure, physico-chemical properties of soil are properties with determinant role in ion exchange process characteristic parameters values in dynamic conditions. Thus, a significant rise of formation time (t_f) value, evolution time (t_e) of mass transfer zone, the cambic chernozem soil factor (f) can be seen compared with sandy soil. Regarding the length of mass transfer zone (L_{ZTM}), the values are closed, and the formation time (t_f) and evolution time (t_e) of mass transfer zone and factor (f) rise significantly with rising of the clay content of soil.

For P sorption rate determination a first order kinetic model was used (Powell et al., 1998):

$$C = C_0 \cdot e^{-kt} \qquad (11)$$

where: C is the P concentration in effluent at time t (mg L^{-1}); C_0 is the initial concentration in feed solution (mg L^{-1}); k is the first order rate constant (h^{-1}); t is time (h).

When the concentration in the effluent reached at half of the initial concentration ($C/C_0 = 0.5$), results of the half time $t_{1/2}$:

$t_{1/2} = (\ln 2)/k$, and the values of k were determined from graph slope $\ln(C/C_0) = -kt$.

Using the values from Table 3 and equation (11) parameters $t_{1/2}$ and k were calculated and the obtained values for the two types of soil are presented in Table 5.

Using directly the data from Table 3 and linear interpolation method it was found that $t_{1/2} \cong 128$ h for cambic chernozem soil, and $t_{1/2} \cong 19.5$ h for sandy soil. When we compared these values with the ones presented in Table 5 we established that data were identical for cambic chernozem soil and somehow different for sandy soil.

After the examination of the data presented in Table 5 it was obvious that the half time is approximately six times higher in the case of cambic chernozem soil compared to the sandy soil probably due, mainly, to clay presence that offers free valences that add phosphate ions, so, an important role is attributed to soil composition (see Tables 1 and 2).

From kinetic data obtained we can deduce that P sorption in soil is a slow process, the fact demonstrated by the relatively low values of rate constant, k = 0.0054 h^{-1} for cambic chernozem soil and k = 0.031 h^{-1} for sandy soil (Powell et al., 1998).

The suitability of the three types of equations (4), (5) and (6) was tested with experimental data from the two soil types presented in Table 3.

Model coefficients estimation was realised by non-linear regression using experimental data.

The calculation programme used was Microcal Origin version 6.0 and the initiation parameters value was in all cases 1, except the case of logistic equation, when the initiation parameter value α, was 0.1 for cambic chernozem soil. The values of coefficients for the three equations and two soil types, and also the corresponding correlation coefficient value R^2 are presented in Table 6.

Table 4. Characteristic parameters of ion exchange process in dynamic conditions

Characteristic parameters	Cambic chernozem soil - Teleorman	Sandy soil -Dabuleni
t_e	210 (h)	74 (h)
t_f	84 (h)	51.65 (h)
f	0.5	0.302
ϕ	0.5	0.3
L_{ZTM}	27.39 (cm)	28.52 (cm)
θ_U	54 (%)	33.63 (%)

Table 5. Kinetic model paramaters

Soil type	$t_{1/2}$ (h)	k (h^{-1})
Cambic chernozem	128.3	0.0054
Sandy	22.43	0.031

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[1] a correction of mass transfer area lenght by introducing the form factor, ϕ (see Table 4)

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Table 6. Coefficients values a, b, n, k, α, β from the three proposed models and corresponding correlation coefficients R²

Soil type	Model Eq.*	χ^2	R² (model)	Coefficients values				R²' (parameters)
Cambic soil	(4)	0.00078	0.99461	b	4.2884E-6	n	2.48979	0.99882
	(5)	0.00260	0.98213	a	217.27637	k	1.3095	0.30151
	(6)	0.00084	0.99422	α	4.00989	β	-0.03176	0.9378
Sandy soil	(4)	0.00159	0.98890	b	0.01173	n	1.42671	0.97704
	(5)	0.01239	0.91361	a	66.71386	k	0.54419	0.20154
	(6)	0.00244	0.98300	α	2.6984	β	-0.1406	0.81902

Model Eq *: 4 = Rosin-Rammler; 5 = Schuhman; 6 = Logistic equation; Correlation coefficients are calculated for model (R²) and for model paramaters (R²')

From the values presented we concluded that Rosin-Rammler models and logistic type equation offer a better aproximation of the breakthrough curves ($R^2 = 0.98 \div 0.99$) for the soil types studied compared with Schuhman model ($R^2 = 0.91 \div 0.98$). In case of Rosin-Rammler model and logistic equation the values of the b, n, α and β coefficients have good physic significancy while in the case of Schuhman model, a and k parameters do not have physical significance due to extremely low value of correlation coefficient ($R^2 = 0.20 \div 0.30$).

As physical significance for b, n, α and β coefficients we can attribute the sense of rate kinetic constant. Knowing that sorption process is defined by an adsorption stage followed by a desorption stage, to n and β is attributed the sense of rate costant for adsorption process and to b and α the sense of costant for desorption process. So, in these conditions the best simulation is obtained after the equation Rosin-Rammler. It can be observed that the adsorption process is more important than the desorption one. The negative value of the paramater β from logistic equation indicates an ireverrsible tendency of the sorption process. This tendency can be caused by a retrogradation of P in forms of low mobility.

The breakthrough curves comparatively with experimental points were drawn using the models with coefficient values evaluated by non-linear regression (Figures 3 and 4).
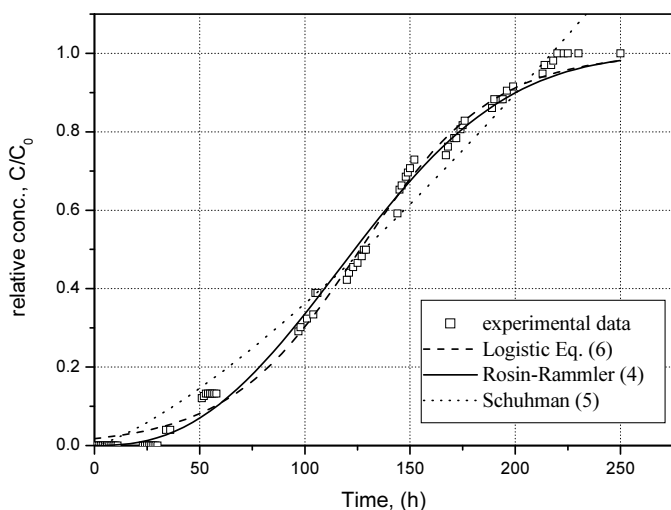

Figure 3. The breakthrough curves expressed using the three models comparatively with experimental points for cambic chernozem soil
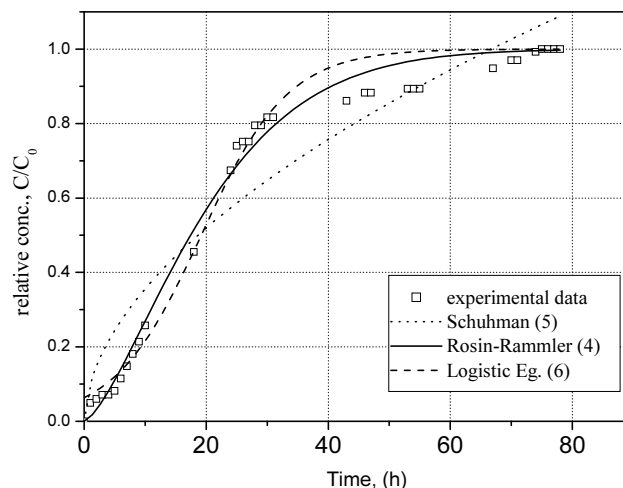

Figure 4. The breakthrough curves expressed using the three models comparatively with experimental points for sandy soil

From the analysis of the breakthrough curves expressed using the three models comparatively with experimental points it can be established that phosphate (P) transport and sorption kinetic on cambic chernozem and sandy soil can be mathematically described with enough precision with Rosin-Rammler model and logistic equation, and from these two methods, the Rosin-Rammler model is better, model in which for the two model paramteres we can ascribe physical significance.

Conclusions

On the basis of experimental research done regarding dynamic phosphate migration regime in a cambic chernozem and sandy soil we can conclude:

– in natural regime, cambic chernozem (Teleorman) has a moderate mobile P supply, and psamosoil (Dabuleni) has low supply with mobile P;

– from kinetic data obtained it can be established that P sorption in soil is a slow process, illustrated by the relatively low values of rate constant of these processes;

– from the research guided in dynamic regime for studying the process of P migration in soil were received information regarding the rate of P sorption in soil that's influenced by the diffuse stages and which can be anticipated with enough

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accuracy by using the testing models involved in describing these processes;

– from the models proposed for testing it was found that Rosin-Rammler model is the most adequate in order to describe the breakthrough curves, and there was obtained a correlation coefficient $R^2 \geq 0.98$;

– cambic chernozem soil due to its high content of organic matter and clay minerals has a better phosphate adsorbtion capacity as compared to with sandy soil;

– the information obtained after these researches are adding to the useful data bank in this field;

– experimentally obtained data presented in this paper are useful in phosphate ion behaviour in soil approach in dynamic conditions and from a stochastic point of view.

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