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SOIL PHYSICS

Quantitative Estimate of the Heterogeneity of Solute Fluxes Using the Dispersivity Length for Mathematical Models of Pesticide Migration in Soils

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Abstract—Mathematical models of solute movement in soils are used as management means for assessing the migration of agrochemicals and calculating environmental risks. In these models, the soil block includes the soil values of dispersivity length or solute diffusivity as one of the main parameters. Under laboratory conditions, this parameter was determined experimentally in soil columns by recording the dynamics of the effluent concentration and solving inverse problems. A direct experimental method of field determination based on the movement of marker solution was tested. For the prediction, risk calculation, and management of pesticide application using physically based mathematical models, the following stepwise procedure is recommended: (1) model parameterization based on experimental soil properties; (2) use of the field dispersivity length, which was 3–11 cm for the 0- to 40-cm layer, exceeded 12 cm for deeper fissured BEL and BT (40- to 60-cm) horizons with prismatic structure, and had a median value of 21 cm in layers below 60 cm in experiments on an agrosoddy-podzolic soil (Eutric Albic Glossic Retisol (Abruptic, Loamic, Aric) (WRB, 2014) from Moscow oblast.

Keywords: soil, solute movement, hydrodynamic dispersion, migration, model PEARL 4, preferential flows
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INTRODUCTION

To prevent the negative consequences of the application of agrochemicals and to calculate the environmental risk, it is important to predict their behavior in natural objects with a specified accuracy, which requires the knowledge of the transit functions of soil. The transit of solutes in the soil is inseparably related to the transport of water; however, the soil is a heterogeneous dispersed porous body composed of a matrix (solid soil particles) and a heterogeneous porous space, which determines specific mechanisms of water movement in the soil. The knowledge of the mechanisms of water movement in pores is a key factor of soil physics, which allows one to determine the leaching rate of solutions from the soil profile, calculate the application rates of preparations, and prevent the contamination of groundwater.

The pore space of soils has a complex structure with water-conductive paths of different sizes and shapes. The classification of pores by sizes is well established [18, 20], but the differentiation of pore shapes also exists and continues to be developed; in particular, an interesting *F* test was suggested by Skvortsova [7]. Ions migrating in large capillaries, so-called preferential migration paths, reach the lower boundary of soil more rapidly than those moving in finer and wavy pores [10]. In addition, the movement rate of solution is maximum in the pore center and minimum at the pore walls, which is related to the effect of soil surface and the presence of a double electric layer hampering the movement of solutions. Therefore, local micro- and mesomixing of solutions takes place in the pore space of soils [10]. Macrostructural phenomena affecting the solution movement also occur in the soil; these are different water permeabilities of layered and fissured soils and sediments and the presence of through cracks and closed pores and voids.

One of the common approaches to the description of water and solute transport in soils is based on the convective-diffusive transport equation, or the so-called differential model of water transport, which relies on the differential Richards equation [8].

Although this model has been used for more than 100 years, it cannot be considered universal for all soil types, especially soils with high contents of clay and organic matter [34]. No uniform stationary front is observed in the real soil. The movement of water and solutes can occur in soil macropores and cracks. A peculiar feature of these migration paths is that most solutes moving with soil solutions have no time to interact with the soil matrix and can rapidly appear in the groundwater [24]. Even substances with different chemical properties are transported by the above mechanism, and migration mainly depends on local soil and climatic conditions [25].

The redistribution of substances in soil solution caused by the complex structure of pore space and the different flow rates in pores of different diameters and shapes is called hydrodynamic dispersion (mechanical dispersion, convective dispersion) D_h. The flow rate in pores v and the soil structure have the highest effect on the D_h value, which is calculated from the equation

$$D_h = \lambda |v|^n,$$

where λ is the coefficient accounting for the dispersion of flow rate in pores of different sizes termed as the parameter of the hydrodynamic dispersion of soil or as the dispersivity length (cm, m); exponent n depends on the degree of soil aggregation. The value of λ can be defined as an elementary mixing cell. It increases with increasing heterogeneity of packing of soil particles [11]. Experiments on the determination of dispersivity length in soils and sediments showed a significant dispersion of this value, from 0.2 cm in sands to several tens of meters and even kilometers during the transfer of solutes in the landscape, e.g., during the migrations of substances in groundwater. However, the mean dispersivity length is 10^{-1} to 10^0 cm for model environments (sand, glass beads, sand–clay mixture), 10^0 to 10^1 cm for bulk sand samples, and about 10^1 cm for soil monoliths [9]. Experiments performed by Aidarov [1] demonstrated that the λ value nonlinearly increases with in heavier-textured soils from 0.6 cm in sands to 50 cm in heavy loams and does not depend on the concentration and filtration rate of solutions. Korsunskaya found that the parameter n varies from 1.20 to 1.35 in chernozems, from 1.17 to 1.60 in soddy-alluvial soils, and from 1.30 to 1.52 in gray forest soils [11]. However, the practical determination of the relationship between the parameter n and the flow rate is time- and labor-consuming; therefore, n is taken equal to 1 in most cases [9, 14].

A review by Vanderborght and Vereecken considered the effect of different factors on the value of dispersivity length [33]. The values of λ are conventionally measured in water-saturated soils. The dispersivity length does not depend on the solution flow rate for coarse-textured soils (sand, loamy sand, sandy loam), while it is directly related to the flow rate for soils of heavier texture. This phenomenon is due to the presence of fine pores in clayey soils, while under increasing hydraulic head, coarser interaggregate pores are involved in the transport, and the dispersivity length also increases [9, 33]. However, note that stable macropores make the major contribution to conductivity at high infiltration of solutions into the soils [17]. If abundant precipitation falls after a long dry period, water bypasses interaggregate pores and follows preferential migration paths. Under unsaturated conditions, the mechanisms of the effect of water content on the dispersion are more complex [22]. In experiments with soil column filled with a sandy loamy soil [26], the dispersivity lengths for the soils with the initial water content typical of the dry season were higher than for the soil in the wet season. Thus, it may be concluded that the soils having preferential migration flows and stable macropores also have higher dispersivity lengths. In the absence of a physically substantiated description of pore space and the corresponding flow characteristics, the value of dispersivity length should be determined experimentally for real soils with a specific organization of pore space. A laboratory experiment with washing of soil columns and solution of inverse problems (CFITIM model [31]) is presently the main method for the experimental determination of this parameter. However, it is perfectly clear that laboratory experiments, even with soil monoliths, cannot reproduce the movement of solutes in the natural soil. Some works point to a direct necessity of such experimental procedures [21]. Therefore, it is important to substantiate and test the direct experimental method of field determination based on the movement of marker solution. Thus, the aim of this work was the field determination of dispersivity length for soddy-podzolic soil and the following testing of the dispersivity length values obtained earlier in laboratory and field experiments during the simulation of pesticide migration in the soil.

OBJECTS AND METHODS

In 2015, a plot experiment (plot size 25 m^2) on the migration of the medium-mobile (K_{oc} = 241 cm^3/g) and medium stable (DT_{50} = 34 days) pesticide cyantraniliprole in an agrosoddy-podzolic soil (Eutric Albic Glossic Retisol (Abruptic, Loamic, Aric, Cutanic) (WRB, 2014), which was described in detail [16], has been launched at the Zelenograd Experimental Station of the Dokuchaev Soil Science Institute in Pushkin district of Moscow oblast [2, 19]. The main properties of soil horizons are given in Table 1. Approximation parameters of soil water retention curve (SWRC) (determined in laboratory by capillarimetry and the method of equilibrium over saturated salt solutions [9]) by the van Genuchten function using the RETC model [32] are given in Table 2.

Soil samples for the determination of residual pesticide were collected by the drilling method 0, 7, 30, 53, and 101 days after the treatment of soil with the pesticide at vertical intervals of 5 cm; two mixed sam-

Table 1. Some physical and chemical properties of agrosoddy-podzolic soils used in the simulation of pesticide transport

Parameter	Horizon			
	PY	EL	BEL	BT
Depths/thickness, cm	0–36/36	36–42/6	42–51/9	51–80/29
Carbon, %	1.10	0.20	0.17	0.22
pH	6.52	6.29	6.07	5.66
Soil density, g/cm^3	1.2	1.35	1.28	1.43
Solid phase density, g/cm^3	2.60	2.78	2.72	2.72
Porosity, cm^3/cm^3	0.51	0.51	0.53	0.47
Texture, %:				
clay, <2 μm	10.6	5.2	10.4	12.7
silt, 2–50 μm	87.9	93.5	89.0	87.3
sand, >50 μm	1.5	1.3	0.6	0.0
Coefficient of filtration, m/day	0.143	0.137	0.428	0.376

ples were then composed from 10 individual samples each, and the content of pesticide in them was determined by liquid chromatography. In addition, temperature loggers were installed on the experimental plot at the depths of 5, 15, and 30 cm, which recorded temperature every hour; the water content was determined in soil layers by thermogravimetry at each sampling time.

Procedure for determining the dispersivity length. We earlier performed a laboratory filtration experiment, the procedure of which is described in the literature [9, 10, 27] and is actively used for experiments with pesticide migration [6], in columns with undisturbed soil monoliths for the determination of dispersivity length [14]. We proposed a procedure for the field determination of dispersivity length based on the observation of marker substance movement. The food dye Brilliant Blue, which is low toxic and low sorbable and shows a good reproducibility of migration paths in the soil, was used as a marker. Before the beginning of the experiment, the soil plot was saturated with water, and tubes 10 cm in height and 5 cm in diameter were installed on the prepared surface and filled with a water solution of the marker substance. After the end of filtration of the coloring solution, vertical sections along the margin and in the center of the tube were exposed for examination. The obtained distribution patterns of marker substance reflected the paths of water movement. We selected five horizontal sections at the of 0, 5, 20, 40, and 60 cm, where the dye experiment was performed in triplicate. The dispersivity length was determined as the difference between the averaged line of Brilliant Blue advance and the boundaries of the forming infiltration flows, which corresponds to the physical mechanism of dispersivity length formation. The morphological separation of the averaged line and the lower boundary of Brilliant Blue penetration under field conditions are schematically presented in Fig. 1. The median values were used in calculations.

Model PEARL 4.4.4 is the latest version from the series of PEARL models, which is developed and actively used in combination with standard scenarios of input data for the prognostic calculations of solute migration in Europe [23]. This model is also applied for predicting the migration of pesticides in Russia, and the All-Russia Research Institute of Phytopathology has developed standard scenarios [4]. Options for considering preferential migration flows appeared in the new version of the model. We used PEARL 4 to simulate the field experiment on pesticide migration and assess the contribution of dispersivity length to the final prediction. PEARL has a block structure, and the movement of water is described by the Soil–Water–Atmosphere–Plant (SWAP) model, in which transport in micropores is added to convective-diffusion transport, and the pore space is divided into two domains: (1) pores throughout the soil profiles (main bypass domain) and (2) pores terminated at different depths (internal catchment domain) [17, 29]. Tiktak et al. analyzed the operation of the new block and the simulation of pesticide migration [29]. They showed that the concentration of pesticide in the flow from the lower boundary of the soil significantly increases, even

Table 2. Parameters of SWRC approximation by the van Genuchten equation

Parameter	Horizon			
	PY	EL	BEL	BT
θs	0.5454	0.5414	0.5424	0.3951
θr	0.0802	0.0784	0.0810	<0.01
α	0.0140	0.0183	0.0110	0.0121
n	1.5235	1.5075	1.5240	1.1315

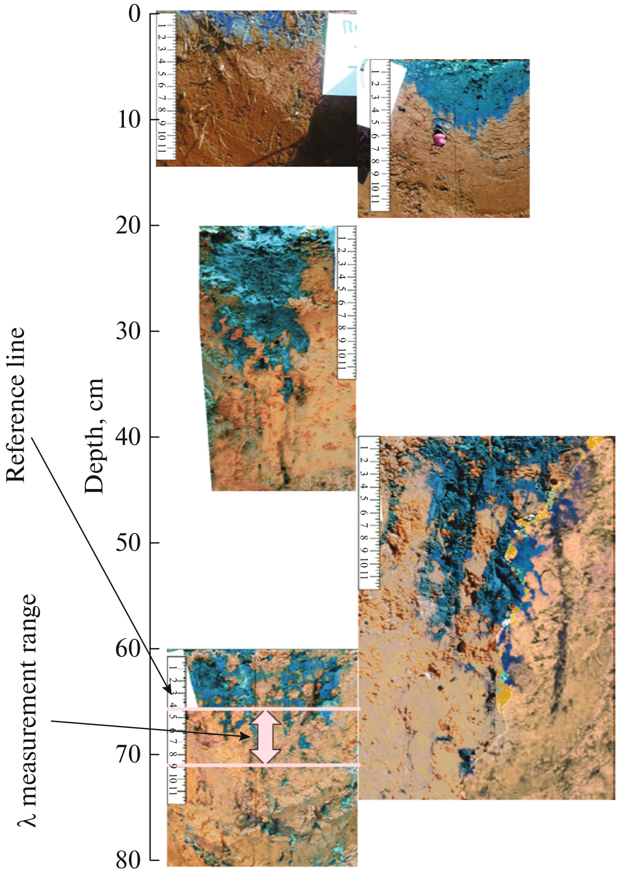

Fig. 1. Migration of marker substance in the soil profile.

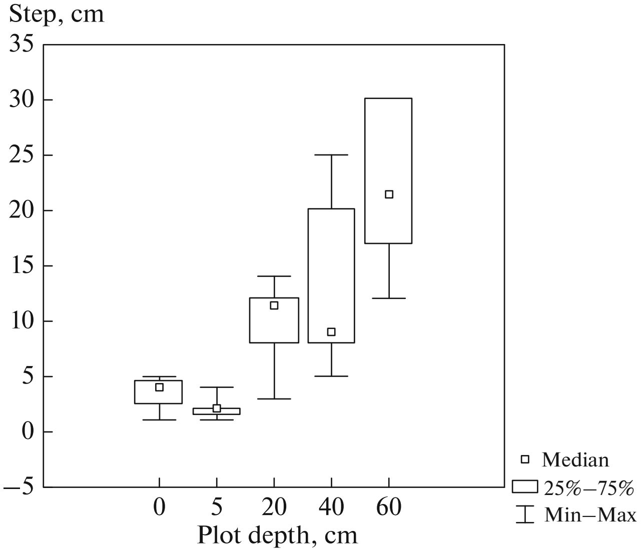

Fig. 2. Statistics of the field determinations of dispersivity length.

under relatively scarce precipitation, primarily due to the migration of solutions through macropores (main bypass domain) and the absence of sorption processes on the surface of the soil solid phase [29] in the course of preferential migration. Unfortunately, the model has a significant limitation: the user cannot parameterize the equations describing the division of pores into macro- and microdomains. In addition, it does not take into account that the pesticide migrating in macropores appears in the outflow significantly earlier and at high concentrations. Therefore, an experimental determination of dispersivity length with due consideration for its natural variation is recommended for specific soils.

RESULTS AND DISCUSSION

Sensitivity analysis of models based on the convective diffusion equation [3, 13, 21, 28] for pesticides showed that the prediction of pesticide in soil runoff depends on four input parameters of the model soil block: density, organic matter content, filtration coefficient, and dispersivity length. The naturally and anthropogenically controlled variations in soil density, organic matter content, and filtration coefficient are studied fairly well. Much less is known about the dispersivity length. This characteristic remains hardly determinable in many aspects within the framework of the experimental supply of models for solute movement in soils. Sensitivity analysis of the PEARL model [12] showed that the front of solute dissolves regularly, when the dispersivity length increases: the more dissolved the pesticide front, the lower its concentration and the earlier it appears at the lower boundary of the profile. An increase in parameter λ is related to the enhanced complexity of the pore space, which results in increasing variation of transfer rates. As a result, the concentration peak of the moving pesticide becomes diffuse. The variation of dispersivity length in the PERL model does not affect the total amount of the substance removed from the profile (the balance law is observed); only the removal time and peak concentrations are changed. These variations of solute movement can be used in the model setting; the use of the dispersivity length values exceeding those recommended in the model PEARL instruction (5 cm) [30] is justified in this case.

Quantification of dispersivity length. Our filtration laboratory experiment in undisturbed monoliths of soddy-podzolic soils 30 cm in height and 10 cm in diameter was described earlier [14]. The obtained value of dispersivity length reaches 40.3 cm.

Let us analyze the field experiment on the determination of dispersivity length (Fig. 1). For the surface and the plot at a depth of 5 cm, the solute front was continuous and had well-defined boundaries. When the depth of the studied plot increased, the front gradually dissolved, and the boundary became tongued. At the depth of 60 cm, it was difficult to determine the averaged penetration line on the vertical section of the profile, because there was no continuous front; instead, there was a network of mottles.

Table 3. Scaled root mean square errors of predicting the residual pesticide contents at different dispersivity lengths (λ, cm) for different layers of agrosoddy-podzolic soil

Depth, cm	λ		
	5 cm (default)	laboratory	field
0–10	0.328	0.33	0.33
10–15	1.42	0.761.412.49	1.571.982.052.9
15–20	1.98	26.440.16	30.16
20–25	2.133.940.16		
25 cm and deeper			
0–60			

From the field data, the following values of dispersivity length were determined: 3–11 cm for the layers of 0 to 40 cm and more than 12 cm for deeper layers with a fissured prismatic structure (BT horizon, 40 to 60 cm). Median values were used for the following simulation (Fig. 2).

Model parameterization. The PEARL model requires knowledge of the initial conditions, those at the lower and upper boundaries, and experimental sources (physical and chemical properties of soils and toxicants). The prognosis was started long before the addition of pesticide to the soil, at the beginning of the year, to avoid the effect of the initial conditions on the prognosis. The parameterization of experimental sources of the soil block was based on the above laboratory and field determinations of the properties of soil horizons (Table 1). Data on the physicochemical properties of the pesticide cyantraniliprole were borrowed from the conclusions of the European Food Safety Authority [19]; however, the reported half-life period (34 days for soils significantly different from the agrosoddy-podzolic soil) was replaced by DT_{50} = 49.9 days, which was found by the All-Russia Research Institute of Phytopathology for the soil type under study [5]. Daily meteorological data obtained using a portable meteorological station under field conditions were used as conditions at the upper soil boundary[1].

After the parameterization of the model with physical soil properties, the model was tested using different values of dispersivity length. Three options were tested: (1) the default value of dispersivity length (5 cm recommended by the designers); (2) the value determined in the laboratory experiment; and (3) the values found in field determinations with a marker. Layer data for each sampling time (0, 7, 15, 53, and 101 days) were compared with the model results. The scaled root mean square error (SRMSE) was calculated for each soil layer from five experiment–model pairs of values (Table 3).

The analysis of the results shows an increase in the predicting capacity y of the model at the dispersivity lengths different from the default values. No appreciable difference was found between the options with the laboratory and field experimental values of dispersivity length, especially for nonfissured aggregated plow horizons. The field morphological determination of dispersivity length, even in a first approximation, gave the results well agreed with the approved and adjusted laboratory filtration method and appreciably increased the prognostic capacity of the models in the case of prismatic soil structures with cracks and macropores. The method proposed for the determination of dispersivity length under field conditions obviously requires versatile testing for soils of different geneses, properties, texture, and consistence.

CONCLUSIONS

Within the framework of the study, the direct experimental method for the field determination of dispersivity length based on the movement of marker solution was tested. Parameterization of the model was done using the experimental data for its soil block (density, texture, SWRC parameters, etc.). The results of modelling of the field experiment on pesticide migration showed an improvement of prognosis at the use of dispersivity length values experimentally determined under field and laboratory conditions. The field method of dispersivity length determination promptly gives experimental values of dispersivity length for soils with different textures, which makes it possible to accurately describe the migration of the pesticide cyantraniliprole and to calculate environmental risks with consideration for the variation of the soil pore space.

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