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Numerical and experimental study of solute transport in unsaturated soils

Stanisław Maciejewski

Polish Academy of Sciences, Institute of Hydroengineering, 80-953 Gdańsk - 5, Kościerska 7, Poland

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ABSTRACT

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The results of a study on the hydrodynamic dispersion of ionic solutes in unsaturated porous media are presented. This study concerns the description of solute transport in moving coordinates, the application of the radiometric method to measure the water content and solute density in unsaturated soil, the verification of the theory of solute transport in the aeration zone. The application of moving coordinates has permitted the elimination of the convection term from the transport equation, which leads to more simple numerical procedures. This is due to the absence of the so-called numerical dispersion, which is present in the classic approach and brings about substantial errors. The radiometric method of monitoring has allowed to simultaneously measure the distribution of water content and solute density in the undisturbed soil sample. The experimental results have shown a linear relationship between the coefficient of dispersion and pore-water velocity for a given water content, and a linear dependence of the dispersion coefficient on the water content for a constant pore-water velocity.

1. INTRODUCTION

Solute transport in unsaturated soils is a problem of hydrodynamics in a multiphase medium. Research on this problem requires a measurement method which allows one to investigate the change of density of separate phases of the medium, i.e. water content (θ) and contaminant density (ρ), during groundwater flow. Such a method, radiometric monitoring, is presented in this paper. The distribution of θ and ρ in time and in space allows one to verify theoretical models of hydrodynamic dispersion.

The research of hydrodynamic dispersion also requires a method of calculation of values of contaminant density from the transport equation. The problem of numerical dispersion in such an equation is a difficult one;

therefore many authors have looked for a method of eliminating this difficulty. One of these methods which avoids numerical dispersion in the solution of the transport equation is presented in this paper.

2. DESCRIPTION OF HYDRODYNAMIC DISPERSION IN MOVING COORDINATES

The one-dimensional transport of solute in an unsaturated porous medium is described by the following equation:

$$\frac{\partial \rho}{\partial t} = \frac{\partial}{\partial t}\left[D\theta \frac{\partial}{\partial x}(\rho/\theta)\right] - \frac{\partial(\rho v_{\mathrm{w}})}{\partial x} \tag{1}$$

where ρ = density of contaminant in porous medium [M L^{-3}]; θ = water content in soil [L^3L^{-3}]; D = coefficient of hydrodynamic dispersion [L^2T^{-1}]; v_{w} = pore-water velocity [LT^{-1}] ($v_{\mathrm{w}} = J_{\mathrm{w}}/\theta$, J_{w} = Darcy velocity); x = space coordinate [L]; and t is time [T].

Instead of the contaminant density ρ one can use the contaminant concentration C. The relationship between ρ and C is described by:

$$\rho = C\theta \tag{2}$$

Using this description we can express Eq. 1 in the following form:

$$\frac{C\theta}{\partial t} = \frac{\partial}{\partial x}\left(D\theta \frac{\partial C}{\partial x}\right) - \frac{\partial(C\theta v_{\mathrm{w}})}{\partial x} \tag{3}$$

Eq. 3 is generally used in recent literature to describe contaminant flow in unsaturated soil (e.g., Warrick et al., 1971; Bresler, 1973a, b; Kirda et al., 1973; Van der Pol et al., 1977; Selim et al., 1977; Smedt and Wierenga, 1978). In this paper the hydrodynamic dispersion equation containing density (Eq. 1) is preferred, as it is connected with the measurement method which allows one to measure the density of the contaminant and not concentration.

The difficulties of a numerical solution of Eq. 1 are connected with numerical dispersion which is caused by the convection term in the transport equation. This problem was discussed by van Genuchten and Wierenga (1974), and by Gaudet et al. (1977). They proposed using a correction term in the coefficient of hydrodynamic dispersion by which the influence of numerical dispersion is decreased. For the one-dimensional problem of transport, the Lagrangian description often gives a good result. The moving coordinate $\xi = x - v_{\mathrm{w}}t$ eliminates the convection term from the hydrodynamic dispersion equation for saturated flow (Scheideger, 1960; Bear, 1972). But this approach does not eliminate the convection term in the hydrodynamic dispersion equation for unsaturated porous media (Eq. 1). Another moving coordinate which eliminates this term in the unsaturated zone is presented by Maciejewski (1985). The moving coordinates of the point (ξ, t) were introduced by the corresponding Euler coordinates (x, t). The relationship between coordinates ξ and x is described by the differential equation (Maciejewski, 1987b):

$$\frac{\partial \xi}{\partial t} = -v_{\mathrm{w}} \frac{\partial \xi}{\partial x} \tag{4}$$

The new functions P, W and d defined in moving coordinates (ξ, t) were introduced as follows,

— contaminant density P in moving coordinates:

$$P(\xi, t) = \rho[x(\xi, t), t] \frac{\partial x(\xi, t)}{\partial \xi} \tag{5}$$

— water content W in moving coordinates:

$$W(\xi, t) = \theta[x(\xi, t), t] \frac{\partial x(\xi, t)}{\partial \xi} \tag{6}$$

— coefficient of hydrodynamic dispersion d in moving coordinates:

$$d(\xi, t) = D[x(\xi, t), t] \left[\frac{\partial x(\xi, t)}{\partial \xi}\right]^{-2} \tag{7}$$

The inverse transformation allows one to determine functions ρ, θ and D all in Euler coordinates from functions P, W and d,

— contaminant density ρ:

$$\rho(x, t) = P[\xi(x, t), t] \frac{\partial \xi(x, t)}{\partial x} \tag{8}$$

— water content θ:

$$\theta(x, t) = W[\xi(x, t), t] \frac{\partial \xi(x, t)}{\partial x} \tag{9}$$

— coefficient of hydrodynamic dispersion D:

$$D(x, t) = d[\xi(x, t), t] \left[\frac{\partial \xi(x, t)}{\partial x}\right]^{-2} \tag{10}$$

Eqs. 5–10 present the relationship between density ρ, water content θ and coefficient of hydrodynamic dispersion D in Euler coordinates and density P, water content W and coefficient of dispersion d in moving coordinates.

Introducing ρ, θ and D (Eqs. 8, 9 and 10, respectively) into Eq. 1 one obtains:

$$\begin{aligned} &\frac{\partial P}{\partial t} \frac{\partial \xi}{\partial x} + P \frac{\partial^2 \xi}{\partial x \partial t} + \frac{\partial \xi}{\partial t} \frac{\partial P}{\partial \xi} \frac{\partial \xi}{\partial x} \\ &\quad = \frac{\partial}{\partial \xi}\left[d W \frac{\partial}{\partial \xi}(P/W)\right] \frac{\partial \xi}{\partial x} - P \frac{\partial}{\partial x}\left(v_{\mathrm{w}} \frac{\partial \xi}{\partial x}\right) - \frac{\partial P}{\partial \xi} v_{\mathrm{w}} \left(\frac{\partial \xi}{\partial x}\right)^2 \end{aligned} \tag{11}$$

Using Eq. 11 and the transformation described by Eq. 4 one can easily obtain the equation of hydrodynamic dispersion described in moving coordinates:

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \xi}\left[dW\frac{\partial}{\partial \xi}(P/W)\right] \tag{12}$$

This equation does not include a convection term. This is important because the numerical solution of Eq. 12 is much simpler than the solution of Eq. 1. The solution of Eq. 1 was obtained indirectly. In the first step from the solution of Eq. 12 the function $P(\xi, t)$ was obtained. In the second step, using Eq. 8, $\rho(x, t)$ was calculated which constitutes a solution of Eq. 1 (Maciejewski, 1987b). The possibility of this method and the comparison between results obtained by this method and a quasi-analytical solution of the infiltration problem presented by Smiles et al. (1978) and Elrick et al. (1979) will be presented in S. Maciejewski (in prep.).

3. METHOD OF MEASUREMENT

Two radiometric methods of measurement were used to study water and contaminant transport in soil: absorption of gamma radiation and the tracer method. The effect of absorption of gamma radiation has been used for measurement of water content in soil, for example by Gurr (1962), Vachaud et al. (1970), and Zaradny (1973). Recently, the absorption method was used by Grismer et al. (1986) for the measurement of contaminant density in unsaturated soil. Tracers are used for observation of the contaminant flow in soil (e.g., Makowski, 1972).

The combination of absorption and tracer methods was applied to the simultaneous measurement of water content and contaminant density in an undisturbed and unsaturated soil sample. The diagram of a measuring position and a measuring apparatus is shown in Fig. 1. The transport of water and contaminant in sandy soil was measured in a Plexiglas® cylindrical column. The length of this column was 75 cm and the diameter 4.4 cm. During the experiment the column was located in a constant-temperature chamber. A soil sample was placed vertically in order to measure capillary rise and infiltration, and horizontally to measure horizontal flow. Water was supplied to the soil sample through a ceramic filter from a Mariotte vessel. The injection of contaminant into the soil sample was made by a syringe through a hole in the wall of the Plexiglas® column. Iodine ions were used as contaminant. The water content was measured by means of a gammascopic method. The radioactive isotope ^{241}Am was used to measure the change in density. The density of the contaminant was measured by means of a radioactive tracer containing the same chemical components as the investigated solute. In this case the

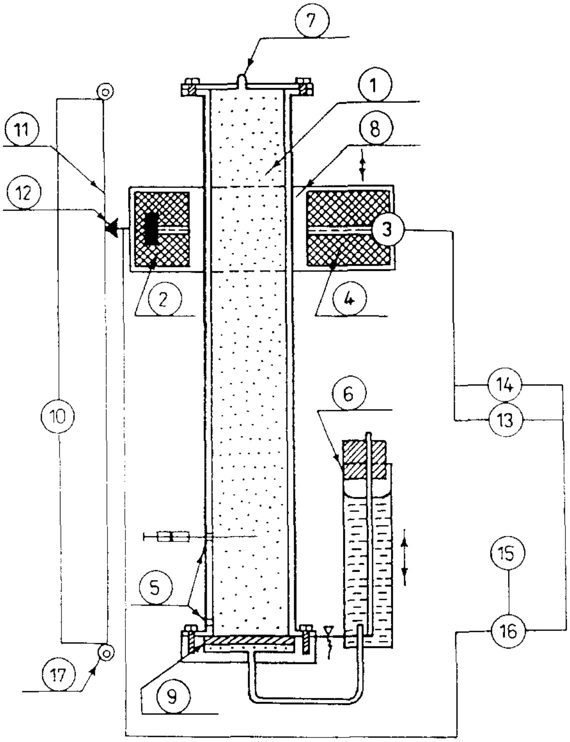

Fig. 1. The diagram of measuring position and measuring apparatus (*1* = soil; *2* = collimator; *3* = detector; *4* = lead screen; *5* = injection holes; *6* = Mariotte water system; *7* = hole from contact with atmosphere; *8* = measuring trolley; *9* = ceramic filter; *10* = low voltage power; *11* = resistance wire; *12* = measuring slide; *13*, *14* = impulse counters; *15* = data logger; *16* = control system; *17* = insulator).

radioactive isotope of iodine (^{131}I) was applied as a tracer of contaminant. The activities of sources of gamma radiation used for experiments were 3.7 GBq for ^{241}Am and 7.4 MBq for ^{131}I. These two radioactive isotopes have different energies of gamma radiation. Two intensities of gamma-ray beams emitted by ^{241}Am and ^{131}I were measured by one detector at different energy levels, i.e. 360 keV for ^{131}I and 60 keV for ^{241}Am. The two measured intensities I_1 and I_2 are described by the following equations (Gurr, 1962; Maciejewski, 1985, 1987a):

$$I_1 = k_1\rho\frac{1-\exp[-L(\mu_g^1\rho_g + \mu_w^1\rho_w + \mu^1\rho)]}{L(\mu_g^1\rho_g + \mu_w^1\rho_w + \mu^1\rho)} + T_1 \tag{13}$$

$$I_2 = I_2^0\exp[-2R(\mu_w^2\rho_w + \mu^2\rho)] + k_2I_1 + T_2 \tag{14}$$

where I_1 = measured intensity of gamma radiation emitted by the radioactive tracer; I_2 = intensity of gamma radiation which is the sum of ^{241}Am radiation (the first term in Eq. 14) and that scattered in soil rays emitted by the radioactive tracer (the second term in Eq. 14); I_2^0 = intensity of gamma radiation emitted by ^{241}Am which reaches the detector when the soil is dry ($\rho_w = 0$); μ_g^i = mass absorption coefficient of gamma radiation for soil (i is the number of energy level) [L^2M^{-1}]; μ_w^i = mass absorption coefficient of gamma radiation for water [L^2M^{-1}]; μ^i =mass absorption coefficient of gamma radiation for the contaminant [L^2M^{-1}]; ρ_g = density of soil [M L^{-3}]; ρ_w = density of water [M L^{-3}]; ρ = density of contaminant [ML^{-3}]; R = radius of cross-section of sample [L]; L = mean effective width of the sample from which the gamma radiation emitted by a radioactive tracer reaches the detector [L]; k_1 = counting yield for the first level of energy [M^{-1} L^3]; k_2 = parameter describing the scattered radiation emitted by a radioactive tracer for the second level of energy; and T_1, T_2 are noise intensities.

The bulk density ρ of each phase (soil, water, contaminant) is defined as $\rho = \mathrm{d}m/\mathrm{d}V$, where m is a mass of a phase contained in a bulk volume of soil V. Eq. 13 describes the absorption of the gamma-ray beam emitted by the radioactive tracer situated inside the soil sample (Maciejewski, 1987a) and Eq. 14 describes the absorption of the gamma-ray beam emitted by ^{241}Am situated outside the sample and transmitted through the soil (Gurr, 1962; Vachaud et al., 1970).

Eqs. 13 and 14 are nonlinear equations where ρ_w and ρ are unknown variables. This set of equations was solved using the Newton method. The solution of the system of Eqs. 13 and 14 for a given measurement point gives the value of the water content θ [$\theta = \rho_w/\rho*$, where $\rho* = 1\,\mathrm{g\,cm^{-3}}$ (= water density)] and the density of the contaminant ρ at this point. Distributions of water content $\theta(x,t)$ and contaminant density $\rho(x,t)$ in time t and space x were obtained by linear interpolation. This method was used for example by Zaradny (1978). The measurement method is described in detail by Maciejewski (1985, 1986, 1987a). The contaminant density profiles in the soil sample for a capillary rise are shown in Fig. 2. The water content profiles for the same experiment are presented in Fig. 3.

These figures do not contain the measurement points because measured values of water content or contaminant density were obtained at different times and in different places. This method of presentation of experimental results is typical for this kind of measurement (e.g., Vachaud et al., 1970; Zaradny, 1978). The estimation of the measurement error was done indirectly by the analysis of the mass balance (contaminant, water) in the soil sample. The ratio r of an average error of the measurement density $\langle\Delta\rho\rangle_x$ to an average density $\langle\rho\rangle_x$ is defined as the relative error of density by the equation:

$$r = \langle\Delta\rho\rangle_x/\langle\rho\rangle_x = (m_m - m_e)/m_e \tag{15}$$

where m_m = measured value of mass obtained by integration; and m_e = exact value of mass measured by the gravimetric method before injection. The

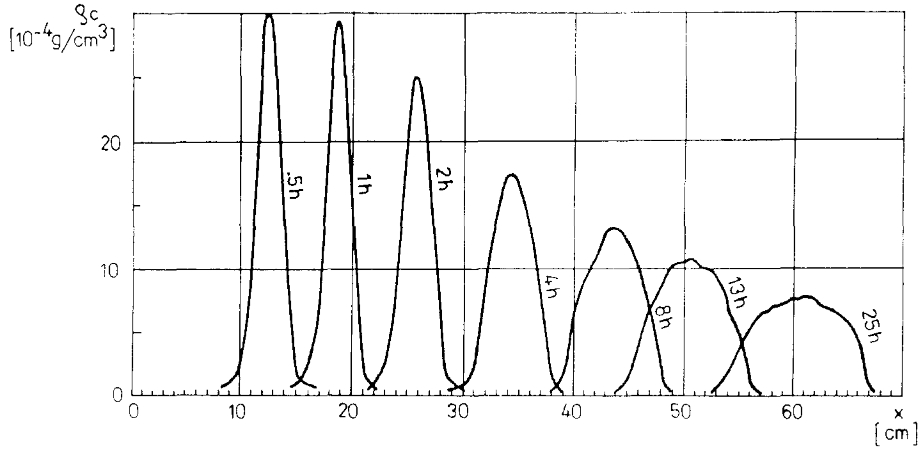

Fig. 2. Distributions of iodine ion density for capillary rise.

averaging is defined in the following way:

$$\langle \rho \rangle_x = L^{-1} \int_0^L \rho(x,t)\mathrm{d}x \tag{16}$$

The histograms of the measurement error r of the contaminant density and water content are shown in Fig. 4. The mean relative error of the contaminant density was ±5.13%. The error in the estimation of water content was obtained in the same way as the error of the contaminant density. The mean relative error of the water content was ±5%. The results of measurement and the analysis of the measurement error proved the suitability of the presented apparatus and method for the study of a solute flow through unsaturated porous media. This method allows one to investigate dynamic

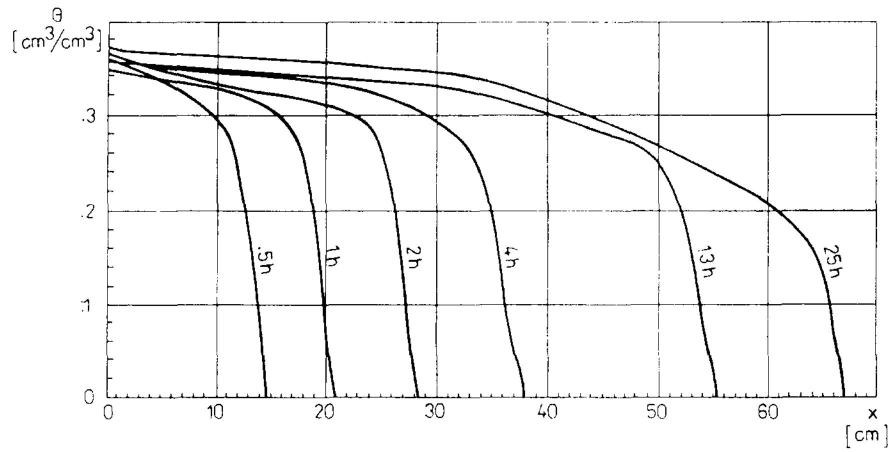

Fig. 3. Distributions of water content for capillary rise.

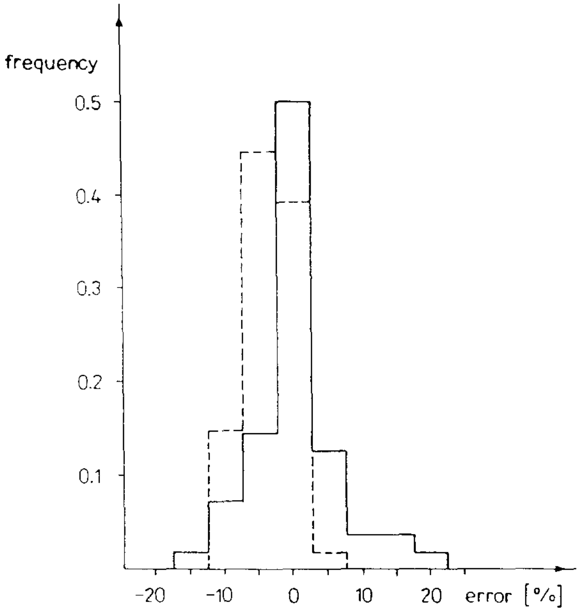

Fig. 4. Histogram of measurement errors (*solid line* = contaminant; *dashed line* = water).

water and contaminant transport in an undisturbed sample. This fact is very important because distributions of water content and contaminant density permit one to verify the theory of solute transport in soil. This method proved to be better than the method presented by Grismer et al. (1986) since it may measure a greater range of contaminant densities, in particular for low density. The advantage of Grismer's method is that it does not use a free source of gamma radiation. The use of a free source of radiation is not an important drawback because activity of gamma rays of the source ^{131}I was during the experiments 7.4 MBq.

4. RELATION BETWEEN HYDRODYNAMIC DISPERSION COEFFICIENT AND WATER VELOCITY AND WATER CONTENT

The experimental distribution of water content and contaminant density permitted the estimation of other parameters, i.e. velocities of water and solute and gradients of contaminant concentration. The velocity of water was determined by integrating two distributions of water content $\theta(x, t_1)$ and $\theta(x, t_2)$ at different times t_1 and t_2. The velocity of contaminant was determined by integrating two distributions of contaminant density, $\rho(x, t_1)$ and $\rho(\mathrm{x}, t_2)$, at different times t_1 and t_2. Water content distribution θ and contaminant density distribution ρ were used to determine the solute concentration C ($C = \rho/\theta$). Fick's law and the above-mentioned parameters were applied to estimate the coefficient of hydrodynamic dispersion. Four experiments were conducted to determine the dispersion coefficient. In order to analyse the variability of the dispersion coefficient as dependent on water velocity, certain water content ranges were used. This analysis was done for the following ranges of water contents: 0.35–0.30, 0.30–0.25, 0.25–0.20, 0.20–0.15 and 0.15–0.10. These d–v_w relationships are shown in Fig. 5. The points indicate experimental coefficients and the straight line shows the approximation. The intercept of this linear approximation was assumed to be equal to the coefficient of molecular diffusion in soil for water velocity equals zero. This relationship can be described by the following equation:

$$D(v, \theta) = \alpha(\theta)v + D_m\tau \tag{17}$$

where α = dispersivity [L]; D_m = molecular diffusion coefficient in the fluid continuum [$L^2\ T^{-1}$]; and τ = coefficient of tortuosity [$L\ L^{-1}$]. The coefficient of tortuosity of the porous medium τ is defined by Bear (1972) as $\tau = (L/L_e)^2$, where L is the length of the segment connecting two points between which fluid flows and L_e is the real length of the path of particles in the porous medium. The definition gives values of the coefficient of tortuosity less than 1. There are other definitions of tortuosity which give values greater than 1 (Carman, 1937).

Considerable scattering of values of the experimental dispersion coefficient is mainly caused by errors of estimate such as gradient of concentration and water and contaminant velocities. Therefore the linear relation was used as a first approximation. The linear relation between dispersion coefficient and water velocity is also valid in saturated media (see Bear, 1972). The linear dependence of the dispersion coefficient on water velocity for unsaturated soils was given, for example, by Gaudet et al. (1977), Melamed et al. (1977),

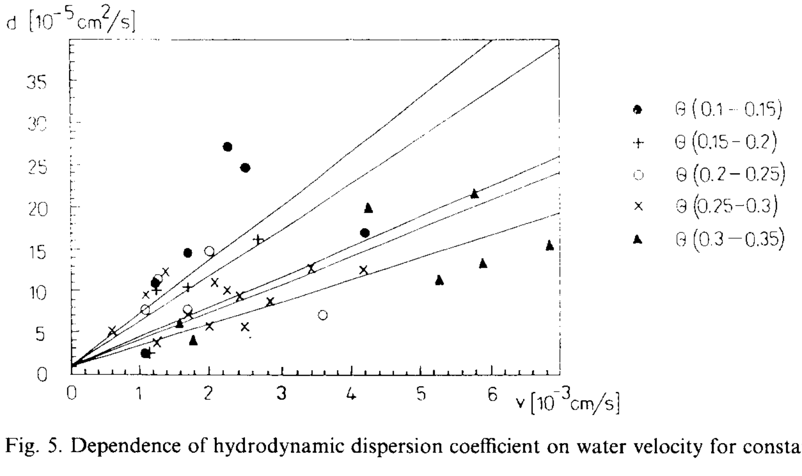

Fig. 5. Dependence of hydrodynamic dispersion coefficient on water velocity for constant water content.

Smedt and Wierenga (1978), and Nielsen et al. (1981). The relationship between dispersivity α and water content θ was investigated. Results of those investigations are shown in Fig. 6 and given by the expression:

$$\alpha(\theta) = -a\theta + b \tag{18}$$

where $a = 0.197$ cm and $b = 0.087$ cm for sandy soil.

This result indicates that a smaller water content gives a greater dispersivity. This fact may be explained by the relation between flow tortuosity and water content. It means that if the water content decreases then flow paths will be longer (increasing tortuosity, i.e. decreasing value of τ) and this will broaden the arrival time distribution which causes higher dispersion.

This dependence of tortuosity on water content follows also from the characteristics of the molecular diffusion in soil. The molecular diffusion coefficient in unsaturated soil D_p described, for example, by Kemper and Schaik (1966) is given by the relation:

$$D_\mathrm{p}(\theta) = D_\mathrm{m} a \exp(b\theta) \tag{19}$$

where D_m = coefficient of molecular diffusion in water; and a, b are parameters of soil.

Since $D_\mathrm{p}(\theta)$ is equal to $D_\mathrm{m}\tau$, the coefficient of tortuosity in unsaturated soil may be described as follows:

$$\tau(\theta) = a \exp(b\theta) \tag{20}$$

Using the definition of this coefficient, it is clear from the above that $L_\mathrm{e}(\theta_1) > L_\mathrm{e}(\theta_2)$ if $\theta_1 < \theta_2$, so both results stemming from Eqs. 18 and 19 agree. It means that for two samples having identical water velocities higher dispersion will be observed for lower water contents. It must, however, be noted that lower water content leads to a smaller diffusion coefficient.

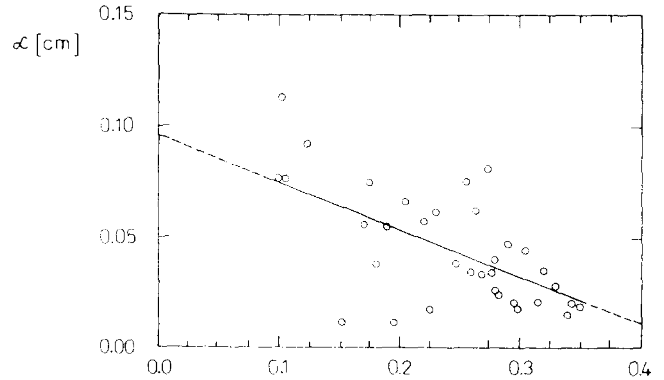

Fig. 6. Dependence of dispersivity on water content.

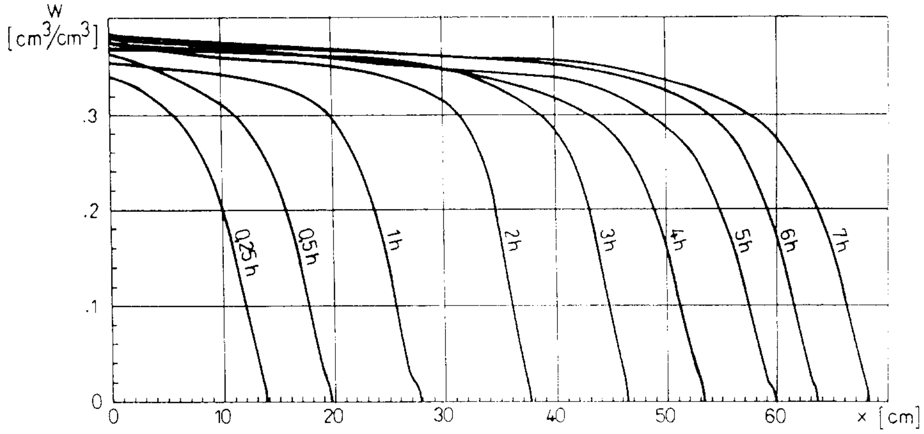

Fig. 7. Measured distribution of water content for horizontal flow.

5. VERIFICATION OF RELATION BETWEEN HYDRODYNAMIC DISPERSION COEFFICIENT AND WATER VELOCITY AND WATER CONTENT

The coefficient of the hydrodynamic dispersion obtained by experiment shows a significant error; therefore the relationship presented in Eqs. 17 and 18 can be treated as a hypothesis. A new series of experiments was conducted to prove this hypothesis. Those experiments concerned the infiltration and horizontal flow. Soil samples at the beginning of the experiments were dry. During experiments the water content and water velocity were highly variable. The measured distribution of water content is shown in Fig. 7, which depicts the results of one experiment. The obtained distribution served then to estimate the water velocity field. Theoretical distributions of

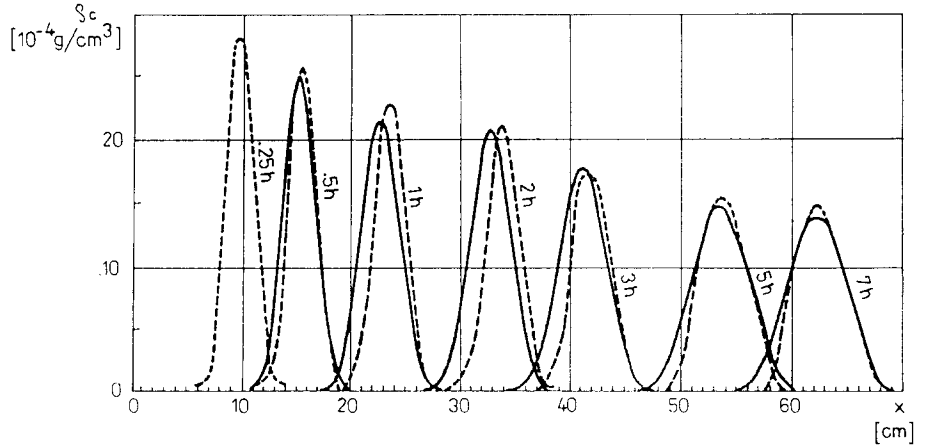

Fig. 8. Comparison between calculated and measured distributions of contaminant densities (*continued lines* = calculated; *dashed lines* = measured).

contaminant density were calculated for those experiments. They were obtained based upon: the model of solute transport in unsaturated soil (Eq. 1), the hydrodynamic dispersion coefficient (Eqs. 17 and 18), the measured distribution of water content and water velocity (e.g., Fig. 7). Moving coordinates were used for solving the transport equation. The comparison of the calculated and measured results of the horizontal flow is shown in Fig. 8. The continuous lines present results of calculations and dashed lines present results of measurements. Statistical methods were used to prove that the set of calculated results is equivalent to the set of measured ones. The comparison of calculated ρ_c and measured ρ_m densities of iodine ions is presented in Fig. 9. The comparison of calculated (x_c) and measured (x_m) positions of the points with maximum densities of iodine ions is shown in Fig. 10. Hence, the statistical analysis of measurements and calculation results proved Eqs. 17 and 18 to be correct.

6. SUMMARY

The radiometric method of measurement of water content and contaminant density may be applied to the study of hydrodynamic dispersion. This method allows a simultaneous and continuous measurement of water content and density of contaminant on intact soil samples. Distributions of these quantities may serve to determine other parameters of hydrodynamic dispersion such as coefficient of dispersion, water and solute velocities, concentration of solute in soil water, and gradient of concentration. The distributions of water content and contaminant density in time and in space may serve to verify theoretical models of hydrodynamic dispersion. The presented results

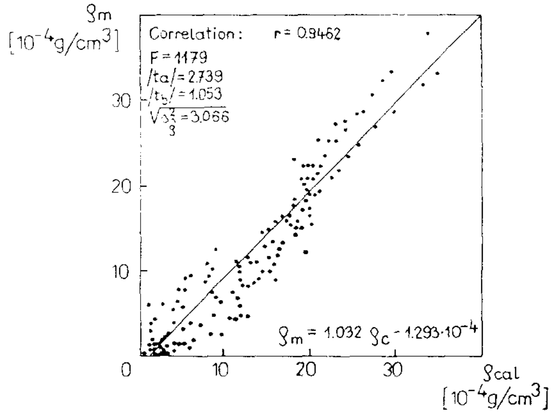

Fig. 9. Relationship between measured (ρ_m) and calculated (ρ) contaminant densities (F = calculated parameter to study linear dependence of ρ_c on ρ_m — test F; t_a, t_b = calculated parameters to study statistical hypothesis H_0: $\rho_m = \rho_m$ — test t; s^2_ρ = residual variance).

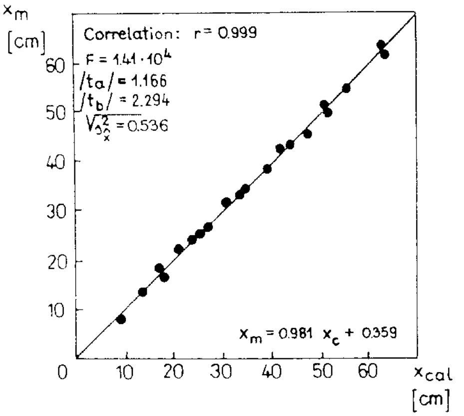

Fig. 10. Relationship between measured (x_m) and calculated (x_c) positions of the points with maximum of contaminant density (F = calculated parameter to study linear dependence of x_c on x_m — test F; t_a, t_b = calculated parameters to study statistical hypothesis H_0: $x_c = x_m$ — test t; s_x^2 = residual variance).

showed a linear relationship between the coefficient of dispersion, water velocity and water content for transport of iodine ions in sandy soil. The relationship between dispersivity and water content was explained by the dependence of tortuosity on water content.

The description of hydrodynamic dispersion in moving coordinates allows to eliminate the errors present in numerical solutions of the transport equation.

The presented methods of measurement and calculation could be used for study of transport of other contaminants in different soils. The solution of the transport equation and the measurement of parameters could be used for prognosis of concentrations of chemical and radioactive substances in the soil.

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