Radioactive tracer techniques used in solute transport studies in a field soil*

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A b s t r a c t. Radioactive tracer techniques were used for in-situ measurements of the solute velocity, longitudinal dispersion coefficient, and bypassing ratio, as well as for estimation of the impact of land-use change on area-averaged nitrate relative concentration profiles. It was found that the impact of land-use on nitrate relative concentration profiles can be studied for small and medium cumulative infiltration only. The mean and dominant velocities obtained from breakthrough curves were significantly higher than those obtained from concentration profiles, but a good agreement of maximal velocities was found. From the results of the bypassing ratio measurement it can be seen that the warm and rainless second half of April 1993 (similar situation repeated in April 2000) resulted in a preferential flow as high in the barley field as that in the untilled soil with well-developed macropore systems. The rain resulting in the macropore flow can appear 24-times on average in south-western Slovakia during the vegetation season. Such rain, combined with the high value of the bypassing ratio can be a cause in the drop of fertilization efficiency, as well as in the shallow groundwater pollution in the Danubian Lowland.

K e y w o r d s: tracer techniques, solute velocity, longitudinal dispersion coefficient, bypassing ratio

INTRODUCTION

If water is applied to the soil surface at rates higher than the infiltration rate into the soil matrix, the surface-vented macropores in field soil can allow water and solutes to pass rapidly through the root zone, bypassing the matrix of the soil [8]. The amount and character of that short-circuiting [2] depends among other things, on tillage practices. Conservation tillage (no-till) yields more continuous macropores and other preferential paths reaching directly from the surface deep into the subsoil. Conventional tillage destroys the structure of the surface soils, mixing the plough layer and covering the macropore's connection to the surface [1]. But a few weeks after tillage, the macropores are formed again as a result of the activity of soil animals (earthworms, ants), the decomposition of plant roots and the desiccation of soil, etc. Conventional tillage may lead to pronounced surface runoff, agri-chemical washout and soil erosion, whereas no-till practices can be a cause of shallow groundwater pollution by agri-chemicals, and a drop in fertilisation and irrigation efficiency.

The aim of this study was to estimate the parameters of water flow and solute transport in soils with and without preferential paths using radioactive tracer techniques.

MATERIALS AND METHODS

Experimental site

The experimental site was located at the Experimental Station of the Research Institute of Irrigation in Most near Bratislava, where small-scale field experiments were conducted between 1993-96. The loam soil is classified as Calcari-mollic Fluvisol [4].

Plots 1.5×3.5 m (1993), 3×3 m (1994), and 1×1 m (1995 and 1996) were used to study the nitrate transport and to estimate the bypassing ratio in that soil. Gamma emitting iodine $^{131}I^-$ was used as a tracer for the nitrate movement in cultivated (1993-95) and untilled (1994 and 1996) soil under spring barley (1993 and 1994), maize (1995), and grass (1994 and 1996).

The radioactive tracer technique

In-situ equipment for radioactive tracer measurements is shown in Fig. 1. The probe consists of a duralumin tube (3) in which a Geiger-Mueller (G-M) detector and analog
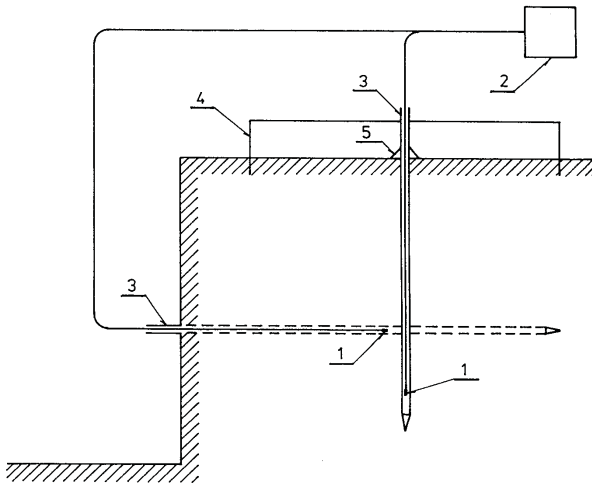
Fig. 1. In-situ equipment for radioactive tracer measurements.

interface unit (1), connected to a nuclear analyser (2) with coaxial cable, can be placed in any position desired. Tubes (10-mm O.D., 8-mm I.D., and 1500-mm length) are inserted ho- rizontally from the pit or vertically from the soil surface into holes made by a 10-mm-diam. steel rod into the soil below the 1-m^2 square infiltrometer (4). Conical soil sealing (5) made for each vertical probe prevents water from bypassing the probe as was proved by a dye test with Methylene Blue. Owing to its small size (21-mm length and 6.3-mm O.D.) the G-M detector can be considered as a point detector [11].

Concentration profile and breakthrough curve measurement

Following tracer pulse application, scanning the vertical probe (Fig. 1) was undertaken to measure the tracer concentration vs depth distributions (concentration profiles) in the course of the infiltration experiment, with a 10-cm distance between the measuring points. The tracer concentration vs time distributions (breakthrough curves) was measured in the horizontal probes (Fig. 1). Both the concentration profiles and breakthrough curves were used to estimate the velocity of the nitrate transport in the soil under study. To differentiate the results, the velocities obtained from the breakthrough curves are marked with an apostrophe. In both cases, the counting rate was considered to be proportional to the tracer concentration [6].

The area-averaged, relative concentration of nitrate in a soil layer can be established along the way as it proceeds. The counting rate in the soil layer from 10a cm to 10(a + 1) cm (for a = 0, 1, 2, ...) was estimated as the mean of counting rates measured by a probe at these two depths. Then the nitrate relative concentration in soil layers surrounding the probe was estimated from the counting rate in the same soil layers so that the sum of the relative concentrations in all layers was equal to 1. Finally, the nitrate relative concen-

tration in the soil layers obtained in a plot were area-averaged according to the method described by Butters and Jury [3], i.e., the area-averaged nitrate relative concentration in a soil layer in a plot is the mean of the relative concentrations obtained in the layer by all the probes.

Estimation of nitrate velocity and longitudinal dispersion coefficient from breakthrough curves

Assuming that shortly after conventional tillage the soil is a homogeneous porous medium, we use the convection-dispersion model to describe a solute transport in it. The dominant, mean, and maximal velocity of nitrate, as well as the longitudinal dispersion coefficient were estimated from the tracer breakthrough curves measured at the depth L in the soil without the surface-vented macropores.

The dominant velocity v_{id}' at the depth L and measuring point i was estimated from the time t_{id} where the maximum in the tracer breakthrough curve was observed:

$$v_{id}` = L/(t_{id} - 0.5\ t_a), \tag{1}$$

where t_a is the time of tracer application.

The mean velocity v_{is}' at the depth L and measuring point i was estimated from the time t_{is} when the half of the tracer applied was transported below this depth:

$$v_{is}` = L/(t_{is} - 0.5\ t_a). \tag{2}$$

The maximal velocity v_{im}' at the depth L and measuring point i was estimated from the time t_{im} of the first detection of the tracer in this depth:

$$v_{im}` = L/(t_{im} - 0.5\ t_a). \tag{3}$$

The mean velocity v_{is}' is a good estimate of the actual distance velocity of nitrate in the soil under study [7] but the measurement of the whole curve would be time consumptive in the case of wide and flat breakthrough curves. In this case, a comparison of the normalised breakthrough curve with model curves (the Sauty's procedure [7]) is used. The normalised breakthrough curve has the maximal concentration equal to 1, minimal concentration equal to 0, and the temporal axis (x-axis) has the same logarithmic scale as the model curves:

$$c_{norm} = [A/t^{1/2}]\ \exp\ [-\ \text{Pe}\ (1 - t)^2/4\ t], \tag{4}$$

where c_{norm} is the normalised concentration, A is a constant chosen to ensure $c_{norm,max} = 1$), t is time, and Pe is the Peclet number.

The model curves for Pe = 1, 3, 10, 30, 100, and 300 are presented by Käss [7]. We chose that model curve by means of which the normalised breakthrough curve was best fitted. From the time t_{id} where the maximum in the tracer breakthrough curve was observed, and the time t_d where the maximum in the model curve was observed, the time $t_{iv} = t_{id}/t_d$ was estimated. From the time t_{iv} the mean velocity v_{is}' = L/t_{iv}

of the nitrate transport in the soil was calculated. Finally, the longitudinal dispersion coefficient D_{il} of nitrate at the depth L and measuring point i was estimated from the Eq. (5):

$$D_{il} = v_{is}' \, L/\text{Pe}. \qquad (5)$$

Estimation of nitrate velocity from concentration profiles

The mean velocity v_{is} of the tracer transport in the measuring point i and time T was calculated from the Eq. (6):

$$v_{is} = z_{is}/(T - 0.5 \, t_a), \qquad (6)$$

where T - time when the tracer concentration profile in the measuring point i was measured, and z_{is} - mean penetration depth of the tracer calculated from the Eq. (7):

$$z_{is} = \sum_{j=1}^{m} z_{ij}\left(n_{ij} - n_0\right) / \sum_{j=1}^{m}\left(n_{ij} - n_0\right), \qquad (7)$$

where z_i - depth in which the counting rate n_{ij} was measured in the measuring point i and time T, n_0 - background counting rate, and m - number of the counting rate measurements in the measuring point i.

The dominant velocity v_{id} of the tracer transport in the measuring point i and time T was calculated from the equation:

$$v_{id} = z_{id}/(T - 0.5 \, t_a), \qquad (8)$$

where z_{id} - depth in which the maximal counting rate was measured in the measuring point i and time T.

The maximal velocity v_{im} of the tracer transport in the measuring point i and time T was calculated from the Eq. (9):

$$v_{im} = z_{im}/(T - 0.5 \, t_a), \qquad (9)$$

where z_{im} is the maximal depth of tracer penetration measured in the measuring point i and time T.

Bypassing ratio measurement

In the soil with surface-vented macropores, the bypass flow mechanism allows more water to flow down immediately to deep layers bypassing the soil matrix [14]. The bypassing ratio BR is defined as a ratio of the rate of bypassing the soil matrix (in this study via macropores and interaggregate pores) to the rate of total flow. The bypassing ratio BR increases with an increase of application rate from $BR=0$ for an application rate less than the infiltration rate into the soil matrix, to the maximal value. BR = max for an application rate higher than the value of saturated hydraulic conductivity of the soil under study [8]. Changes in crack parameters have to be taken into account in modelling the bypass flow in a swelling-shrinking soil [12].

The bypassing ratio was measured with the radioactive tracer technique, too. The leading edge of radioactive tracer ^{131}I was formed in the soil matrix below the area of maximum plant uptake, and its displacement h was measured with the G-M detector placed in the tube inserted vertically into the soil (Figs 1 and 2). The bypassing ratio was calculated from the Eq. (10):

$$BR = 1 - hn_{ef}/I, \qquad (10)$$

where I is the cumulative infiltration of water resulting in the displacement h of the radioactive tracer leading edge formed in the soil matrix, $n_{ef} = \Theta_s - \Theta_r$ is the effective porosity of the soil under study, Θ_s is the saturated water content, and Θ_r is the residual water content of the soil matrix [10].
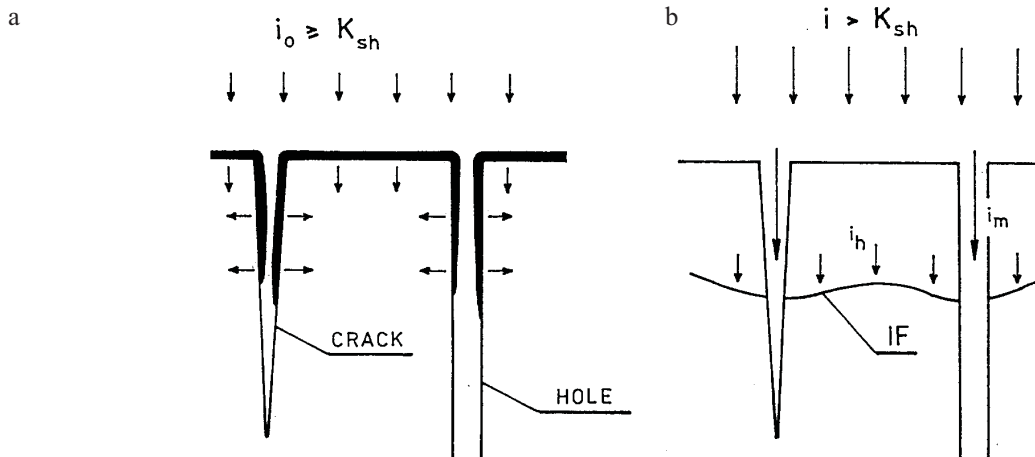

Fig. 2. Two-domain flow of water in macroporous soil: a) infiltration of tagged water into the soil matrix from the soil surface and from macropores (crack, earthworm/ant hole) and interaggregate pores; b) movement of the leading edge of the radioactive iodide IF in the soil matrix at the rate i_h, induced by sprinkled irrigation with the intensity $i > K_{sh}$.

RESULTS AND DISCUSSION

The tracer penetration depths measured with vertical probes in the 1994 field experiment were checked off with those simulated by a numerical model. It was found that neglecting the effects of macropore flow can be the cause of underestimation of the position of the infiltration and re-distribution front with the models based on the Richards' equation for a homogeneous soil [13].

From a comparison of the velocities obtained from the breakthrough curves and concentration profiles measured in the 1993-94 small-scale field experiments, a good agre-ement of maximal velocities was found. A statistical treat-ment of 43 pairs of measurements gave the results as follows (mean ± standard error): $v_m = (8.03 \pm 3.21) \times 10^{-6}$ m s^{-1}, $v_m` = (10.79 \pm 5.09) \times 10^{-6}$ m s$^{-1}$, and $v_m` = 3.233 + 0.912\, v_m$, r = 0.581. As to the mean and dominant velocity, the velocities obtained from breakthrough curves are significantly higher than those obtained from concentration profiles ($v_s' \approx 1.5\, v_s$, $v_d' \approx 2\, v_d$). A statistical treatment of 28 pairs of measu-rements gave the results: $v_d = (2.11 \pm 1.78) \times 10^{-6}$ m s^{-1}, $v_d` = (5.62 \pm 3.72) \times 10^{-6}$ m s$^{-1}$, and $v_d` = 2.495 + 1.485\, v_d$, r = 0.713 for the dominant velocities, and: $v_s = (2.79 \pm 1.47) \times 10^{-6}$ m s^{-1}, $v_s` = (4.41 \pm 3.14) \times 10^{-6}$ m s$^{-1}$, and $v_s` = 0.199 + 1.509\, v_s$, r = 0.708 for the mean velocities [9]. A statistical treatment of 28 pairs of measurements gave the results (mean ± standard error): $D_l = (1.32 \pm 0.94) \times 10^{-7}$ m^2 s^{-1} for the longitudinal dispersion coefficient.

The impact of land-use change on the area-averaged nitrate relative concentration profiles was also studied du-ring the 1993-96 experiments performed in Most. The expe-riments have shown that the impact of land use on the tracer relative concentration vs depth distributions was significant for small and medium cumulative infiltration ($I = 40$-54 mm) but not so significant for the larger cumulative in-filtration ($I = 100$-108 mm). It seems that there exists a threshold in cumulative infiltration (similar to the Lagran-gean length scale for diffusion-type process [5]) required for any particle transported with liquid to have a chance to move through all pore-size categories [11].

As for the bypassing ratio measurement performed in Most in 1993-96, it was found that the preferential flow was about 49% of the total flow ($BR = 0.49$) as measured at the depth of $z = 48$ cm in a spring barley field in April - May 1993, $BR = 0.15$ in the depth of $z = 56$ cm in a maize field in July 1995, $BR = 0.55$ in the depth of $z = 78$ cm in the grassland soil in August 1996. It can be seen that the warm and rainless second half of April 1993 (a similar situation was repeated in April 2000) resulted in the formation of cracks and a preferential flow as high in the barley field as in untilled soil with well-developed macropore systems. As calculated by Faško [11] from observations lasting over 10 years, the rain resulting in the macropore flow can appear 24-times on the average in south-western Slovakia during the vegetation season. Such rain, in combination with the high value of the bypassing ratio can be a cause in the drop of fertilization efficiency, as well as of the shallow ground-water pollution in this region [11].

CONCLUSIONS

Ease and speed of measurement are the most significant advantages of the radioactive tracer technique. One point in a concentration vs depth distributions requires one minute only, installation or removal about 10 min per probe. When used in a field soil, the radioactive tracer technique is non destructive and able to locate heterogeneities. The dose of radioactive tracer necessary for one plot is very small and in the case of $^{131}I^-$ it is one sixth of the dose used in thyroid gland therapy.

The tracer transport parameters (velocity, longitudinal dispersion coefficient) serve to predict fertiliser transport in the soil under study and the calibration of prediction models. The bypassing ratio can serve as an indicator of the drop in irrigation and fertilisation efficiency, as well as in shallow groundwater pollution.

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