A nuclear tracer technique for investigation of solute transport in the unsaturated zone

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Abstract A nuclear tracer technique is presented as a promising tool for detailed investigation of solute transport in the unsaturated zone of field soil during small-scale field experiments. Several gamma-emitting nuclides and their various compounds are suitable tracers used for simulation of transport behaviour of solutes with different mobility (depending on adsorption, exchange processes, etc.) in field soils. The measurements are rapid and applicable to evaluating spatial variability in solute movement with minimal disturbance. The results can serve for evaluation of groundwater pollution from waste disposals or from large-scale agricultural production, and for demarcation of protection zones around water resources.

INTRODUCTION

Several investigations of solute transport in various field soils show that water does not always infiltrate the soil uniformly (piston flow), but it often shows a preference for certain pathways. In a macropore soil, solute bypasses the matrix (bypass flow), or laterally infiltrates from macropore into the matrix (internal catchment) (Beven & Germann, 1982). Fingering of water and solutes is formed in a dry sandy soil or in a stratified soil (Steenhuis & Parlange, 1991), funnelling in a soil with inclined layers (Kung, 1990). These unsaturated zone processes can allow surface-applied agricultural chemicals and leachates from waste disposals to move rapidly down to groundwater, posing problems of environmental hazard.

Because of the ease and speed of in situ measurements, nuclear tracer techniques were used to follow the transport of solutes in the unsaturated zone of soils. Gamma-emitting nuclides with detectable energy of photons and with appropriate half-life of radioactive decay, e.g. $^{51}$Cr, $^{54}$Mn, $^{58}$Co, $^{65}$Zn, $^{82}$Br, $^{115}$Cd, $^{131}$I, $^{137}$Cs, $^{203}$Hg, $^{210}$Pb, etc., and their various compounds are suitable tracers used for the simulation of transport behaviour of solutes with different mobility (as a function of adsorption, exchange processes, etc.) in field soils.

MATERIALS AND METHODS

The probe (Figs 1(a), 3(b)), by means of which the transport of a selected tracer in the unsaturated zone of soil can be monitored (Lichner, 1985), consists of duralumin tube 3 (inner diameter of 8 mm, outer diameter of 12 mm), in which a Geiger-Mueller (G-M)
detector (with a length of 21 mm and diameter of 6.3 mm) and analog interface unit 1, connected to the nuclear analyser 2 with coaxial cable, can be lowered to any desired depth. The probes are inserted into the holes (with a depth of 1.5 m and diameter of 12 mm) made with a steel rod before (infiltration without ponding) or after infiltration (infiltration with ponding). The counting rate-depth \( n = [f(z)]_T \) and counting rate-time \( n = [f(t)]_T \) relationships can be measured with this device.

Since counting rate \( n \) can be considered to be proportional to the tracer concentration (IAEA, 1975, p. 78), the relationship \( n/n_0 = [f(I)]_L \), where \( I \) is cumulative water input, for selected radioactive tracer measured with the detector can take the place of the normalized breakthrough curve \( c/c_0 = [f(t)]_L \) of a solute with similar transport behaviour in the reference depth \( L \) for steady-state flow of water (infiltration rate \( i_0 = \) constant) and negligible evapotranspiration, and the relationship \( n/n_0 = [f(z)]_T \) can take the place of the solute normalized concentration distribution \( c/c_0 = [f(z)]_T \).

Because the velocity of the horizontal movement of water and tracer could be greater than that of the vertical movement in dry soil (Yeh et al., 1985), the soils were wetted before tracer application. In accordance with Jury’s transfer function model (Jury, 1982) one can then assume that at any particular location the tracer velocity is constant with depth and the spreading of a pulse (hydrodynamic dispersion) is attributed to the horizontal variability in vertical transport velocities (convective velocity fluctuations about the mean solute velocity).

There are several ways to use the measured data for the assessment of solute transport. Trudgill et al. (1983) used the depth \( z_p \) of tracer penetration (Fig. 1(b)) as a measure of the effect of macropores upon infiltration. This depth can serve to calculate the maximal velocity of solute in field soil and time of the first appearance of solute in groundwater.

![Fig. 1 Solute velocity measurement during infiltration with ponding. (a) Measuring device arrangement. 1 - G-M detector and analog interface unit, 2 - nuclear analyzer (scaler), 3 - probe, 4 - double-ring infiltrometer, \( \times \) - position of concentration peak. (b) Results obtained (dashed line = background counting rate).](image)

The tracer velocities \( v_i \), \( v_{i*} \), and \( v_{im} \) can be calculated from the arrival time \( t_{im} \) of peak concentration using the breakthrough curve measured at the point \( i \) at a reference depth \( L \) (Biggar & Nielsen, 1976):

\[
v_i = \frac{L}{(t_{im} - 0.5t_a)}
\]
from the depth $z_{im}$ of peak concentration in a tracer distribution at the point $i$:

$$v_{im} = \frac{z_{im}}{(t_i + t_{i-1} + 0.5t_i - 0.5t_a)}$$

(2)

as well as from the mean value $z_i^*$ of tracer penetration depth at the point $i$:

$$v_i^* = \frac{z_i^*}{(t_i + t_{i-1} + 0.5t_i - 0.5t_a)}$$

(3)

where $t_i$ — duration of infiltration, $t_a$ — solute application time ($t_a \approx t_i$), $t_i$ — duration of the tracer distribution measurement at the point $i$, $t_{i-1}$ — duration of the tracer distribution measurements at previous points, and $z_i^*$ — the mean value of tracer penetration depth at the point $i$ which can be found from:

$$z_i^* = \frac{\sum_{j=1}^{m} z_{ij}(n_{ij} - n_b)}{\sum_{j=1}^{m} (n_{ij} - n_b)}$$

(4)

where $z_{ij}$ is depth in which the counting rate $n_{ij}$ was measured at the point $i$, $n_b$ is counting rate of the background (dashed line in Fig. 1(b)), and $m$ is the total number of counting rate measurements at the point $i$.

In the case of infiltration with ponding (Fig. 1), the first 1 cm layer of water in the inner ring of the double-ring infiltrometer 4 is tagged with radioactive tracer and transported by the water passing through the soil. After steady-state velocity $v_s$ is reached, the infiltration experiment is finished. Subsequently, the probes are inserted into the soil, and the tracer distribution (i.e. the relationship between the counting rate $n$ and the depth $z$ — Fig. 1(b)) is measured in the soil inside (positions 1-5, Fig. 2) and outside the inner ring of the infiltrometer. In the case of field soil, the tracer distributions in several positions inside the inner ring of the infiltrometer are different from each other (Fig. 2). However, outside the inner ring no tracer is present. In that case, the tracer velocity $v_i^*$ at a point $i$ inside the inner ring of the infiltrometer can be found from equations (3) and (4).

The presence of a tracer outside the tagged area is evidence of a horizontal component of the water flow velocity (e.g. in the more permeable layer of a stratified soil).

In the case of infiltration without ponding, the nuclear tracer technique was used to measure the breakthrough curve of a tracer at the reference depth $L$ and the tracer distribution in soil.

Around each of eight probes inserted into the clay loam soil to the depth of 1.5 m (Fig. 3(a)), the narrow pulse of Na$^{131}$I solution with the activity of about 10 MBq (0.27 mCi) was trickled by syringe into the annulus with the diameter of about 10 cm, and the counting rates $n_0$ (at time $t = 0$ and depth $z = 0$) were measured in all eight probes. Then the G-M detectors were lowered to the reference depth $L = 30$ cm, and the counting rates $n$ were measured as a function of cumulative water input $I$ while the pulse was leached by sprinkler irrigation over the area of 1.4 m × 3.4 m (Fig. 3(a), dashed line) with infiltration rate $i_0 = 1$ cm h$^{-1}$. Simultaneously, counting rates $n$ as a
function of depth for cumulative water input $I = 4$ cm, 10 cm, and 20 cm were measured. Finally, the velocities $v_i$, $v_{im}$, and $v_i^*$ were calculated using equations (1)-(4).

**RESULTS AND DISCUSSION**

The results of $^{131}$I transport in loamy soil measured in the 35-cm inner ring of the infiltrometer are shown in Fig. 2. The mean values of tracer penetration depth $z_i^{*} - z_5^{*}$ (equation (4)) are 32.1 cm, 17.2 cm, 10.0 cm, 20.6 cm, and 37.3 cm, respectively. The tracer velocities $v_i^* - v_5^*$ (equation (3)) are $2.17 \times 10^{-5} \text{ m s}^{-1}$, $1.10 \times 10^{-5} \text{ m s}^{-1}$, $6.06 \times 10^{-6} \text{ m s}^{-1}$, and $1.73 \times 10^{-6} \text{ m s}^{-1}$, respectively. The measured cumulative infiltration rates $I_i$ are 4.1, 6.3, 9.5, 15.2, and 24.8, respectively.
10^{-6} \text{ m s}^{-1}, 1.19 \times 10^{-5} \text{ m s}^{-1}, \text{ and } 2.05 \times 10^{-5} \text{ m s}^{-1}, \text{ respectively. These results are not typical. In most cases the differences in penetration depth and in velocity are not so high.}

The results of $^{131}$I transport, obtained in clay loam soil during infiltration without ponding, are shown in Figs 4 and 5. Spatial variability of $^{131}$I transport behaviour in that soil, with extremely different results at point 6 (probably due to a clay lens near the soil surface), can be found in both figures. The normalized breakthrough curves for positions 1 - 8 (Fig. 3(a)), measured at the reference depth $L = 30 \text{ cm}$, are presented in Fig. 4. Normalized $^{131}$I concentration distribution for positions 1 - 8 and for cumulative water input of 20 cm are shown in Fig. 5. Similar distributions for bromide and for cumulative water input of 3 cm and 5.8 cm were presented by Germann et al. (1984) and Bowman (1984), respectively. In this study, the velocities $v_i$ and $v_{im}$ obtained from the tracer distribution are approximately the same but they are about one half of the $v_i$ obtained from the arrival time $t_{im}$ of the peak concentration in the breakthrough curve. However,
the velocities calculated from the mean arrival time $t_i^*$ obtained from the breakthrough curve correspond well to the velocities obtained from the tracer distribution. Similarly, in rainfall-runoff modelling, as one parameter of the transfer function, the time difference of the first statistical moments (arithmetical means) of the rainfall and runoff hydrographs is used, instead of their peaks (Nash, 1957; Svoboda, 1987). The first moment of the experimental data is also used in the direct determination of the mean residence time in process vessels in chemical reaction engineering where similar phenomena (e.g. dead space, channelling) and relationships are met as those in field soils (IAEA, 1975, pp. 67-81).

From the shape of measured breakthrough curves and concentration distributions it is obvious that specifications of the peak concentration arrival time $t_{im}$ (e.g. in the point 8, Fig. 4) and the depth $z_{im}$ of maximal tracer concentration could be difficult and ambiguous to establish. Therefore, some continuous curve (e.g. of Fickian, lognormal or gamma function) should be fitted to the measured points and from that curve the specifications of the above-mentioned values would be evaluated.

The area-averaged normalized breakthrough curve can also serve to calibrate the transfer function model (TFM) and a solution of the convection-dispersion equation (CDE) in the way presented by Butters & Jury (1989). The TFM and CDE can then be used to predict the area-averaged normalized tracer concentration distribution for the plot under study.

In the second run of measurements made at the same field on four plots with 36 positions the displacement of the faster peak of $^{131}$I two-peak distribution during redistribution (Fig. 6) was observed. Also registered was the upward movement of $^{131}$I due to evaporation (Fig. 7).

Similar experiments with $^{60}$Co$^{2+}$, $[^{60}$Co-EDTA$]^{-}$, $[^{60}$Co(CN)$_6$]$^{3-}$, and $^{115}$Cd$^{2+}$ were made in clay loam soil where the tracers were injected at the depth of 30 cm with a special injector. In the case of $[^{60}$Co(CN)$_6$]$^{3-}$ one witnessed the incorporation of the ligands CN$^{-}$ into organic compounds resulting in retardation of that tracer. The
The upward movement of $^{131}$I due to evaporation.

\[ \left[ ^{60}\text{Co}(\text{CN})_6\right]^{2-} \text{ velocity } v_m = 4.02 \times 10^{-11} \text{ m s}^{-1} \text{ was the same as that of } ^{60}\text{Co}^{2+} \text{ in comparison with the } \left[ ^{60}\text{Co-EDTA} \right] \text{ velocity } v_m = 1.21 \times 10^{-10} \text{ m s}^{-1}. \text{ The } ^{115}\text{Cd}^{2+} \text{ velocity, measured under different conditions, was } v_m = 3.09 \times 10^{-9} \text{ m s}^{-1}. \]

**CONCLUSIONS**

The nuclear tracer technique is rapid, non-destructive, able to locate the heterogeneities in field soil, and does not influence solute transport. The duration of the measurement of each point in a breakthrough curve or in tracer concentration distribution was only one minute. Installation or removal required about 10 minutes per probe. The time reduction is profitable in the studies of spatial variability of transport properties in the field soils where an extensive set of values is required for statistical processing. The dose of radioactive tracer necessary for one measurement is very small and in the case of $^{131}$I it is one sixth of the dose for thyroid gland therapy.

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**REFERENCES**


