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Penetration and Distribution of Tritiated Water in Soils of a Lysimeter

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A lysimeter experiment was carried out to determine the penetration and distribution of a water pulse applied on the soil surface as a tritium labelled simulated precipitation (30-60 mm). Three plots of soil (each 10 m^2, 1.5 m deep) were used, one filled with gravel, the others with fine sand. In gravel the pulse penetrated ca. 50 cm in the first week, in the fine sand plots less than 15 cm. In gravel the tritium maximum penetrated the lysimeter in 150 days, in fine sand much later. The results were compared with theoretically calculated values.

Introduction

The purpose of this study is to determine the migration and distribution of tritiated water in different types of soil in a lysimeter (Kaitera and Maasilta 1970). The experiment is a joint programme of the Department of Radiochemistry, University of Helsinki, the National Board of Water, Finland and the Helsinki University of Technology as well as the Department of Physical Chemistry and Radiology, Eötvös University, Budapest.

From the hydrological point of view there are two interesting questions:

1) How does the water infiltrate when the types of soil are different and the amounts of water are equal?

2) How does the water infiltrate when the types of soil are equal and the amounts of water are different?

These questions give an idea of the interrelation between precipitation and the formation of ground water which varies rather much not only from place to place but also from one large area to another depending on geological and climatological factors.

The climatological factors and the mechanical characteristics of the soil determine the dynamics of the soil water in the unsaturated zone above the ground water. In fine-grained soils capillary phenomena, diffusion and adhesive forces influence the behaviour of the infiltrating water in different manners.

With a given rainfall the ratio of evaporation to percolation decides the amount of ground water formed. This ratio is different in different parts of Finland and varies with the time of the year.

Tritium has proved to be an ideal indicator when studying the dynamics of soil water as a part of the hydrological cycle (Drost 1978). In the natural environment water containing tritium behaves almost exactly like ordinary water.

The tritiated water formed during an atomic bomb test has been used as hydrological tracer in the 1950-ies already. But few such studies have yet been performed in geographical environments like that in Finland.

From a radiochemical point of view the retention time of tritiated water in different types of soil should be known. This has a great effect on the uptake of tritium by plants and vegetables growing on the contaminated soil. We have tried to answer these questions by measuring the tritium content of the upper soil layer as a function of depth and time, the movement of the maximal tritium content, and by measurements of the total tritium content of the soil profiles.

A mathematical model has been developed to allow us to describe the movement of tritium in the soil, and to increase our understanding of the basic physicochemical processes governing this movement.

Movement of Tritiated Water in Soil

The water movement in soil is mainly due to gravity and to capillary forces. Other effects include temperature gradient, precipitation, evapotranspiration, irrigation and ground-water table.

The movements of the soil water also depend on the degree of saturation of the soil. If the soil is far from saturation, an appreciable part of the infiltrating water is taken up by the soil pores because of adhesive phenomena and only when the saturation deficit has been filled will the ground water formation begin.

It is possible to find the geohydrological relation between precipitation and ground water formation by measuring tritium in water samples from the unsaturated zone. The propagation of a tritium pulse in the soil layers illustrates the movements of the soil water and how these are influenced by evaporation, diffusion and soil frost.

In winter, when the soil is frozen, the tritium pulse will stop moving and the hydrological sequence is broken. In the stage of formation the frost layer attracts water from lower zones, the movement of the soil water is temporarily reversed (Soveri and Varjo 1977) and the ground-water table sinks. This phenomenon was also observed in the present experiments. The melting of snow and frost in spring increases the soil moisture and the level of the ground water table rises.

In summer evaporation normally dries the soil surface. Because of this the moisture gradient rises and the ground-water table sinks. Summer rains stay usually in the upper layer of soil. During autumn evaporation decreases significantly and practically ends in November. Therefore, soil moisture increases and the ground-water table rises.

Climate During the Present Study

If we compare the climatic data of the experiment period and the data of a normal year we can notice that precipitation in July 1977 was 50% greater, in August 40% smaller, in September 10%, October 15%, November 80% greater than average. The thickness of the snow cover was small in February 1978, the average temperature was – 11.1°C. The frost layer of the soil was very thick (60-80 cm) and stabilized quite a lot of water. The spring of 1978 was very dry and the precipitation in April 1978 was 40% and in May 1978 90% smaller than average.

Methods

Field experiments were started on 21 June, 1977, in Otaniemi, a windy summer day, 17°C, on three experimental plots of the Laboratory of Water Resources, Department of Civil Engineering, Helsinki University of Technology. The plots – gravel and fine sand – were chosen to represent the types of soil important to ground-water supply in Finland. They all were without covering plants. Fine sand is a common soil in Hungary also.

The experimental plots – lysimeters – (each is 10 m^2 and 1.5 m deep) were specially prepared, that is artificially filled with different kinds of soil (Fig. 1) (Kaitera and Maasilta 1970). For this experiment one plot of gravel and two plots of fine sand (all without plants) were chosen. A water-collecting system was built under the plots to measure the subsurface flow. The moisture of the soil was measured by neutron-scattering.

The plots were labelled once with tritiated water which was applied directly on the soil using an ordinary garden sprinkler (Table 1). After labelling the plots were irrigated several times by a known quantity of water (Table 2), the first, one hour after labelling.

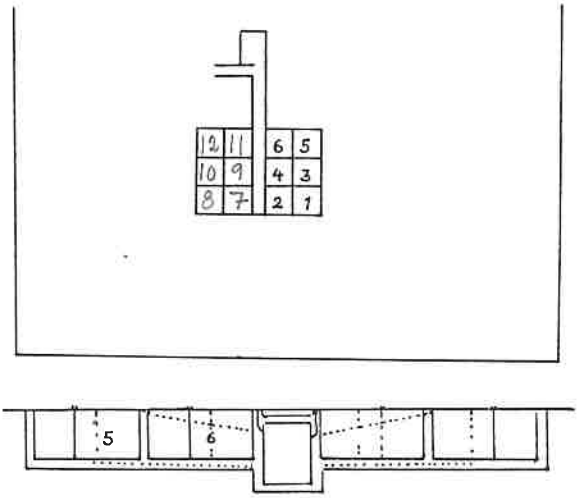


Fig. 1. Lysimeter profiles. No. 3 fine sand, No. 5 gravel and No. 6 fine sand.

We started the experiment by irrigating 30 mm to the plots 3 and 5 and 60 mm to plot 6. These kinds of rain occur in Finland once in 2 years and once in 50 years, respectively. During the following 17 days we irrigated 40 mm to the plot 3, 54 mm to plot 5, and 36 mm to plot 6. At the same time the amount of precipitation was 40 mm.

Soil sampling was started one day after the labelling. The sampling was carried out in the following way: soil cores (two parallels) were taken by a special device (with 2.6 cm of diameter) at every 15 cm from the surface to 135 cm depth. The device was pushed into the soil; after each 15 cm of penetration it was withdrawn and the soil was placed into a glass vessel which was closed and weighed. Finally the hole was filled with the same kind of soil. Parallel to the soil sampling water samples were taken of the water leaving the lysimeter bottom (subsurface flow).

The tritium activity of water from the soil cores taken from different depths has enabled us to determine the distribution of tritium in vertical profiles. Samples taken below the ground-water table are inaccurate because of water movement in the core hole.

Table 1 – Test plots (10 m^2) and application (on June 21, 1977) of tritiated water.

Number of plot	Soil	Amount of water l/m^2 or mm	Amount of activity MBq/m^2 (mCi/m^2)	Specific activity $MBq/(m^2 \cdot l)$ [$mCi/(m^2 \cdot l)$]
3	Fine sand	4.0	65.12 (1.76)	16.28 (0.440)
5	Gravel	4.0	56.98 (1.54)	14.24 (0.385)
6	Fine sand	4.0	65.12 (1.76)	16.28 (0.440)

Table 2– Precipitation and the amount of irrigation water. a)

Days after labelling	Date	Precipitation mm	Amount of irrigation water, mm Lysimeter		
			No. 3	No. 5	No. 6
0	21.6.1977	0	29.7	33.7	57.0
1		0	4.4	5.0	4.9
2		0	7.9	9.4	9.0
3		0.4	–	–	–
4		0	–	–	–
5		2.2	–	–	–
6		0.5	–	–	–
7		0.4	5.3	6,4	6.1
8		0	4.4	5,0	4.8
9	30.6.1977	3.9	3.8	4,7	4.0
10		0	1,0	1,0	1.0
11		4.3	–	–	–
12	3.7.1977	22.0	–	–	–
13		2.6	–	–	–
14		1.1	–	–	–
15		2.6	–	1.0	–
16		0	–	–	–
17	8.7.1977	0	12.7	21.4	6.6
		40.0	69.2	87.6	93.4

The first irrigation was applicated 1 hour after labelling and during 2 hours period.
The amount of water in litres can be calculated by multiplying the amount in mm by factor 10.

b)

Date	Precipitation,			Irrigation water, mm Lysimeter			Subsurface flow, mm Lysimeter		
	mm	Maximum mm	Date of maximum	No. 3	No. 5	No. 6	No. 3	No. 5	No. 6
1.6 –15.6 .77	15.2	12.2	4.6 .77					9.8	
16.6 –30.6	19.1	8.1	19.6	55.5	64.2	85.8		51.0	
1.7 –15.7	49.4	22.0	3.7	25.0	24.4	25.0		52.7	23.2
16.7 –31.7	41.8	12.9	24.7					27.8	21.0
1.8 –15.8	7.4	1.6	14.8						
16.8 –31.8	36.8	11.8	28.8					17.1	
1.9 –15.9	52.4	20.8	12.9					25.3	0.6
16.9 –30.9	20.8	13.1	30.9					2.2	
1.10–15.10	52.7	14.0	1.10				19.4	54.0	65.4
16.10–31.10	27.9	8.8	23.10				29.1	29.5	40.3
1.11–15.11	61.7	14.6	4.11				50.4	51.8	77.8
16.11–30.11	52.3	16.5	24.11				36.4	44.5	45.4
1.12–15.12	9.0	3.1	16.12						
16.12–31.12	24.1	11.2	24.12						
1.1 –15.1 .78	12.9	4.2	1.1 .78						
16.1 –31.1	5.5	3.3	26.1						
1.2 –15.2	1.6	0.8	12.2						
16.2 –28.2	6.4	2.2	28.2						
1.3 –15.3	35.9	8.2	6.3						
16.3 –31.3	17.5	5.6	17.3						0.2
1.4 –15.4	10.2	7.0	15.4					38.1	
16.4 –30.4	15.1	12.5	23.4				1.7	15.5	3.5
1.5 –15.5	2.5	2.3	15.5						
16.5 –31.5	0.5	0.4	18.5	22.4		24.0			
∑	578.7			102.9	88.6	134.8	137.0	419.3	277.2

54 mm was measured as the maximal equivalent of snow on 22.3.1978.

The total amount of tritium in each sample was determined by measuring the amount and activity of the soil water (Environmental Protection Agency 1975). The amount of water in soil cores was determined by weighing the soil samples before and after drying them at 108°C. Two parallel methods were used to extract the soil water: a) isotope dilution, b) freeze-drying. In the isotope dilution method a known amount of water (about 100 g) was added to the soil sample, mixed and after several hours filtered. In the freeze-drying method the soil sample was placed into a vessel of a vacuum distillation apparatus and was frozen by a mixture of ethanol and dry ice. Having evacuated the system and deep-cooled the receiver the water was distilled into the receiver by heating of the vessel containing the soil sample. Activity of the water was measured by a liquid scintillation counter (LKB-Wallac 81000) using 8 cm^3 water and the same amount of Insta-Gel scintillation solution. The samples from the isotope dilution method were measured without distillation, the quenching effect of the salt content – measured by the external standardization method – was found to be negligible. The tritium content of the subsurface flow water was also measured.

The error of the procedure was ~ ± 25% without the error of the labelling.

Experimental Soil Profiles

The gravel profile No. 5 (Table 3) showed that the activity as a function of depth decreased continuously during the first day after labelling. On the second day, the maximum activity was between 30-45 cm. It moved to 45-60 cm on the eighth day (Fig. 2). On the twelfth day after labelling a heavy rain (22 mm) washed most of the activity into the ground water. After 150 days the total activity of tritium between 0-135 cm was no more than 100 times the background. The activity of water collected under the plot reached its maximal value (Fig. 3). The remaining small radioactivity migrated backwards during the winter period and the distribution of the activity became approximately homogeneous in the whole profile (330 days, Fig. 2).

As a function of time, tritium activity decreased after labelling rapidly in the layer 0-15 cm (Fig. 4). In layers 30-45 and 45-60 cm, the activities reached the maximum after 2 and 8 days, respectively. This corresponds to a mean daily advance of infiltration of 16 cm at the beginning and 6 cm at the end of the experiment. The loss of activity calculated from the sum of the tritium activities (Table 3) and the original activity of the sprinkled water (Table 1) was about 40%. The same value was calculated from the original activity of the sprinkled water (Table 1) and the cumulative tritium flux of the subsurface flow of the gravel.

The subsurface flow did not contain significant tritium activity until the beginning of August (Fig. 3). The activity of water began to increase in the middle of August (50 days after labelling), and the maximum level was found on 10 Novem-

Table 3–Activity of tritium, dpm per core, in gravel (No. 5)

cm	1 d	2 d	6 d	8 d	13 d	90 d	150 d	330 d
0– 15	$\boxed{412189}$	68112	22681	7511	8008	4135	7457	7065
15– 30	305868	329275	163966	55715	11194	3668	5096	6477
30– 45	180502	$\boxed{346425}$	$\boxed{270543}$	286670	5450	2678	5120	7686
45– 60	125146	156367	265432	$\boxed{416876}$	21297	3671	2872	9803
60– 75	32346	42411	72447	100912	25353	17160	2972	6476
75– 90	8025	10500	70596	34572	241361	26571	2048	6762
90–105	6011	---	45991	76133	328213	75505	1121	5190
105–120	---	---	---	69982	$\boxed{468540}$	92105	2364	5859
120–135	---	---	---	---	---	$\boxed{207785}$		7283
$\sum$	$1070 \cdot 10^3$	$953 \cdot 10^3$	$912 \cdot 10^3$	$1048 \cdot 10^3$	$1109 \cdot 10^3$	$433 \cdot 10^3$	$29 \cdot 10^3$	$63 \cdot 10^3$
kBq	17.83	15.88	15.20	17.47	18.48	7.22	0.48	1.05
(μCi)	(0.48)	(0.42)	(0.41)	(0.47)	(0.49)	(0.19)	(0.01)	(0.02)

Ground-water table >————<

ber, 1977 (143 days after labelling). The ground-water table was 60 cm below the surface (Fig. 4, Table 3) during the first 150 days.

In the fine sand plot No. 3 (Fig. 5, Table 4) tritium activity decreased with depth during the first twenty days, e.g. the maximum of total activity first remained between 0-15 cm and then moved down very slowly. After 121 and 150 days the maximum was found at 30-45 and 75-90 cm, respectively. After 150 days 90% of the total activity of the whole profile was between 60 and 135 cm, approximately continuously distributed. The total activity of the column shows large variation due to the irregularity of the samples. Evidently, some areas were more efficiently wetted with the labelling solution than other ones.

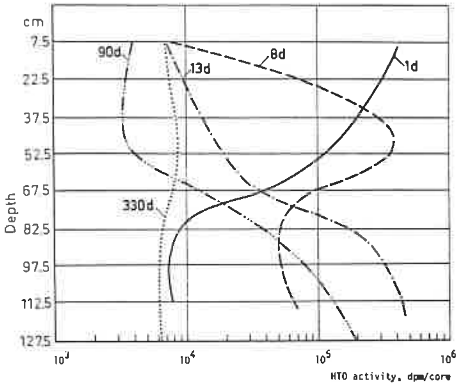


Fig. 2. Tritium activity (dpm/core) of soil water in gravel (No. 5) as a function of depth.

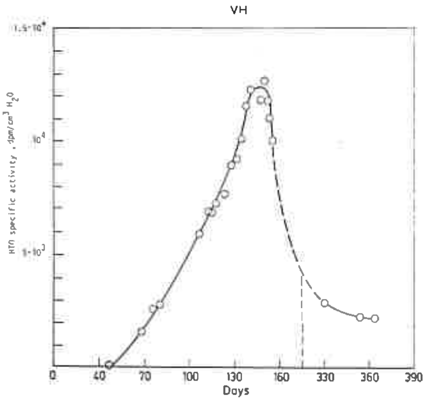


Fig. 3. Specific activity of the water flow through the lysimeter; gravel (No. 5).

Table 4 – Activity of tritium, dpm per core, in fine sand (No. 3)

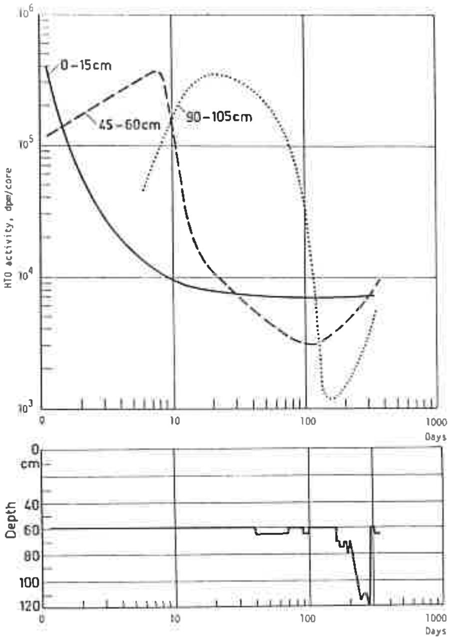
cm	1 d	2 d	8 d	13 d	20 d	34 d	90 d	121 d	150 d	330 d	351 d	370 d
0– 15	1335441	696576	603228	128346	613132	25920	46441	18535	9672	10929	23557	51096
15– 30	164996	128692	201811	607597	183549	762995	246452	49343	5071	7366	45117	154330
30– 45	6287	10887	3477	21312	480309	391829	201218	306307	4356	48146	92596	105916
45– 60	4435	9312	6372	9625	27932		47593	282240	65821	88706	131160	154022
60– 75	4500	– – –	4080	empty	18433	180390	9491	209306	317143	233275	230076	131582
75– 90	– – –	– – –	– – –	– – –	8939	64379	4145	154873	386175	192987	384115	154550
90–105	– – –	– – –	– – –	– – –	– – –	24907	7133	167856	321066	81435	133724	76087
105–120	– – –	– – –	– – –	– – –	– – –	15000	9867	134719	355209	97318	empty	empty
120–135	– – –	– – –	– – –	– – –	– – –	3000	4021	89405	274504	67628	empty	85020
$\sum$	$1516 \cdot 10^3$	$845 \cdot 10^3$	$819 \cdot 10^3$	$767 \cdot 10^3$	$1332 \cdot 10^3$	$1468 \cdot 10^3$	$576 \cdot 10^3$	$1412 \cdot 10^3$	$1739 \cdot 10^3$	$827 \cdot 10^3$	$1040 \cdot 10^3$	$913 \cdot 10^3$
kBq	25.27	14.08	13.65	12.78	22.20	24.47	9.60	23.53	28.98	13.78	17.33	15.22
(μCi)	(0.68)	(0.38)	(0.36)	(0.34)	(0.60)	(0.66)	(0.25)	(0.63)	(0.78)	(0.37)	(0.46)	(0.41)

Ground-water table >———<

Table 5 – Activity of tritium, dpm per core, in fine sand (No. 6)

cm	1d	2 d	8 d	13 d	20 d	34 d	48 d	90 d	121 d	150 d	330 d	351 d	370 d
0– 15	1123218	553414	67041	42089	32054	7282	22390	3961	276	772	8776	13565	45292
15– 30	156881	10268	105519	empty	304869	69867	239652	118105	2184	1238	9332	26358	103303
30– 45	59173	14601	375476	537513	532210	685564	219093	217067	52506	1199	3087	93297	135038
45– 60	39916	9535	empty	14681	405600		664547	389979	218446	1864	24776	98670	230520
60– 75	26008	– – –	8187	13238	71388	282128	22359	162623	402464	10850	58861	275798	232584
75– 90	13015	– – –	3118	– – –	31098	327688	107654	67718	182695	133033	188761	340954	253274
90–105	empty	– – –	– – –	– – –	– – –	125945	32049	108125	174896	305576	371139	316617	empty
105–120	– – –	– – –	– – –	– – –	– – –	empty	– – –	78817	203018	272359	368508	244153	empty
120–135	– – –	– – –	– – –	– – –	– – –	– – –	– – –	35684	126649	338669	310464	141348	153119
$\sum$	1418·10³	578·10³	559·10³	607·10³	1377·10³	1498·10³	1308·10³	1182·10³	1363·10³	1065·10³	1344·10³	1551·10³	1152·10³
*k*Bq	23.63	9.63	9.32	10.11	22.95	24.97	21.80	19.70	22.72	17.75	22.40	25.85	19.20
(µCi)	(0.63)	(0.26)	(0.25)	(0.27)	(0.62)	(0.67)	(0.58)	(0.53)	(0.61)	(0.47)	(0.60)	(0.69)	(0.52)

Ground-water table ⟶



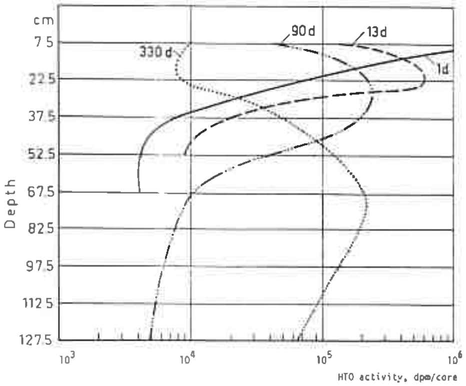


Fig. 4. Tritium activity (dpm/core) of soil water in gravel (No. 5) as a function of time after labelling. Depth of ground-water table.

Fig. 5. Tritium activity (dpm/core) of soil water in fine sand (No. 3) as a function of depth.

The activity migrated backwards during the winter period and showed higher values in the upper layers. This migration continued during the very dry period of May, 1978.

The upward movement of the activity in winter time is caused by the soil frost. As the soil freezes the water is moving toward the freezing zone for thermodynamical reasons. The similar phenomenon later on in May is caused by the abundant evapotranspiration.

The total activity of tritium as a function of time after labelling (Fig. 6) decreased in layer 0-15 cm very slowly but a heavy rain in late July washed the tritium to deeper layers. In layers 15-30, 45-60 and 60-75 cm maximum activities were reached after 34, 121 and 150 days,respectively. Correspondingly, the velocity of infiltration is 0.5 cm per day. The subsurface flow of fine sand No. 3 had no tritium activity until June 1978 (370 days after labelling) when the ground-water table was 135 cm below the surface.

In the fine sand plot No. 6 (Fig. 7, Table 5) the tritium activity decreased with depth continuously on the first and second day after labelling, e.g. the maximum activity was between 0-15 cm during 2 days and moved then downwards with a greater velocity than in the fine sand No. 3 (Fig. 5). Between 8 and 20 days the

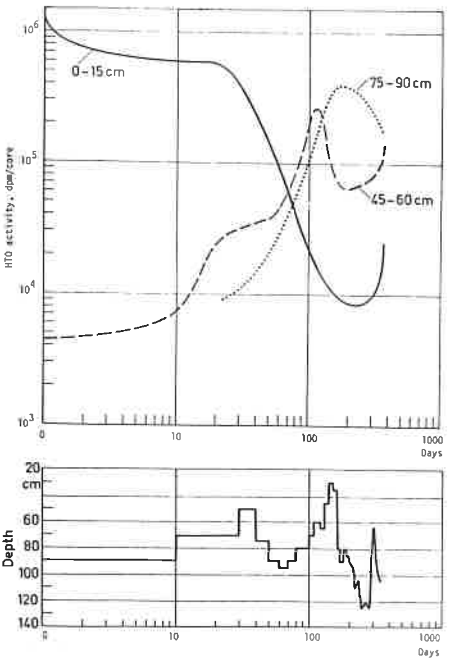


Fig. 6. Tritium activity (dpm/core) of soil water in fine sand (No. 3) as a function of time after labelling. Depth of ground-water table.

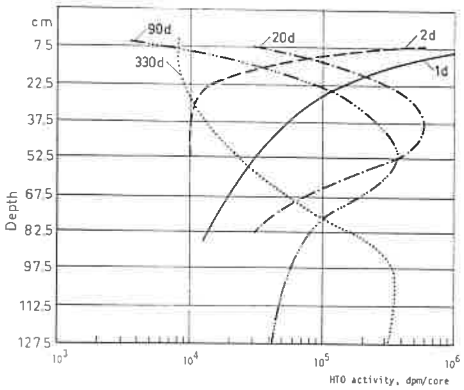


Fig. 7. Tritium activity (dpm/core) in soil water in fine sand (No. 6) as a function of depth.

maximum was at 30-45 cm, between 34 and 90 days at 45-60 cm, after 121 days at 60-75 cm; 95% of the total activity of the whole profile was between 90 and 135 cm after 150 days. Here the velocity of infiltration varied between 2.1 and 0.5 cm per day.

Plot No. 6 was irrigated by twice the amount of water of plot No. 3 after labelling and the differences in activity profiles and velocities are evidently due to this fact and to the differences of the ground-water tables. In plot No. 3 the ground-water table occasionally rose up to 30 cm below the soil surface. In plot No. 6 it rose maximally to 60 cm below the surface (Fig. 8).

The total amount of irrigation water used was ca. 30% higher on plot No. 6 than on plot No. 3 by the end of the experiment and this difference was eliminated practically by the 1 year precipitation. This explains why the velocity of infiltration was the same by the end of the experiment on the two fine sand plots.

The total activity of tritium as a function of time after labelling (Fig. 8) showed the same difference between the two fine sand profiles as showed the maximal activity as a function of depth.

The subsurface flow of the fine sand No. 6 did not contain significant tritium activity until June, 1978 (350 dpm/cm^3) after 370 days).

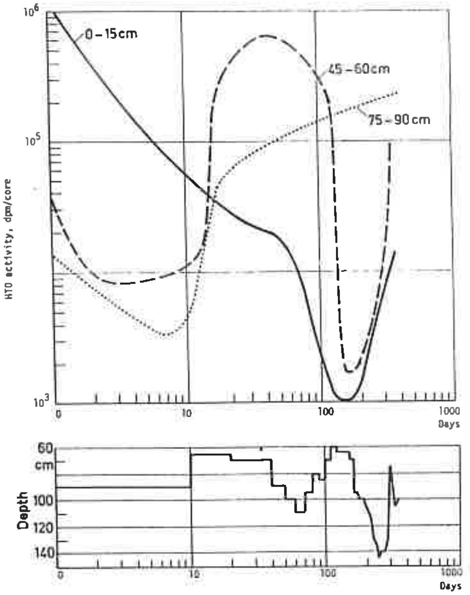


Fig. 8. Tritium activity (dpm/core) of soil water in fine sand (No. 6) as a function of time after labelling. Depth of ground-water table.

In fine sand plots many holes were found during the experiment which caused great variance to the results, as seen when comparing the sum of the activities of the columns. For example, the values of plot No. 3 belonging to 48 days after labelling had to be cancelled because of such irregularities. The loss of activity calculated similarly as in the gravel (Tables 4, 5 and 1) was about 30% in both fine sand plots.

Mathematical Theory

To fit the experimental points a mathematical theory has been developed with analytical solution of the one-dimensional fundamental differential equation describing the movement of diffusing substances. The situation we have is drawn in Fig. 9. The equation mentioned, taking into account the molecular diffusion, mechanical dispersion appearing as a result of turbulence, the moving velocity of the tritium impulse owing to the presence of gravity and capillary forces, absorption and radioactive decay, can be written as

$$\frac{\partial c}{\partial \tau} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - \lambda c \qquad (1)$$

where

$$\tau = \frac{t}{1+\rho K_d/\varepsilon}\,(s)$$

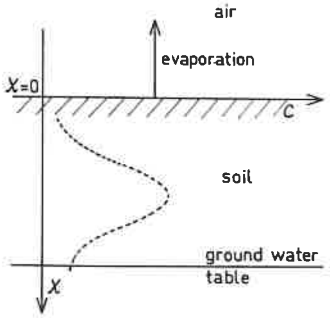


Fig. 9. Coordinate system of the mathematical theory.

t = time (s)
ρ = soil density (g/cm^3)
ε = soil porosity (–)
K_d = equilibrium distribution coefficient (cm^3/g)
c = specific activity of tritium (Bq/cm^3)
x = depth of the lysimeter (cm)
D = diffusion coefficient including longitudinal turbulence (cm^2/s)
v = ground-water movement speed (cm/s)
λ = decay constant (l/s)

To fit the experimental points several solutions of Eq. (1) have been analyzed, with different initial and boundary conditions:

a) for infinite medium:
  - instantaneous source initial conditions,
  - step function source initial conditions,

b) for semi-infinite medium:
  - instantaneous source initial and evaporation boundary conditions at $x = 0$,
  - step function source initial and impermeable boundary conditions at $x = 0$.

Analysis of these solutions showed that after 1 day the initial conditions have little effect, but the boundary conditions at $x = 0$ seriously influence the results. This enables us to use the simpler instantaneous initial conditions with mass transfer (evaporation) boundary conditions at $x = 0$, that is

$$\begin{aligned} &c(\tau=0; \quad x \gtrless 0) = 0 \\ &c(\tau=0; \quad x=0) = \infty \\ &F(\tau>0; \quad x=0) = -\alpha[c(\tau, x=0) - c_s] \end{aligned} \qquad (2)$$

where

F = tritium flux [Bq/(cm$^2 \cdot s$)]; $\alpha \geqslant 0$

α = mass transfer coefficient (cm/s)
c_s = specific activity of tritium in the air (Bq/cm^3)

We seek the solution of Eq. (1) using an equivalent process. This could be a sum of two impulses (c_1 and c_2, with initial activity inventory N_1 and N_2) travelling in an infinite long rod of cross section S in opposite directions from point $x = 0$. Utilizing the well known solutions for a single impulse in an infinite medium (Crank 1957) we get

$$c(\tau,x) = c_1(\tau,x) + c_2(\tau,x) = \frac{e^{-\lambda t}}{S\sqrt{4\pi D\tau}}[N_1\exp(-\frac{(x-v\tau)^2}{4D\tau}) + + N_2\exp(-\frac{(x+v\tau)^2}{4D\tau})] \tag{3}$$

In the following expressions for simplicity the radioactive decay will be neglegted. Utilizing boundary conditions (2) with

$$F(\tau,x) = -D\frac{\partial c_1}{\partial x} + c_1 v - D\frac{\partial c_2}{\partial x} - c_2 v$$

and neglecting c_s we obtain

$$N_2 = N_1\frac{v+2\alpha}{v-2\alpha} \tag{4}$$

The total amount of activity in the region $x > 0$ (in the soil) after sufficiently long time utilizing (3) is

$$N_{\text{tot}}^{+} = S\int_0^\infty c\,dx = N_1$$

and the cumulative amount of activity crossing the boundary at $x = 0$ after sufficiently long time

$$M = S\int_0^\infty F(\tau,x=0)d\tau = -S\int_0^\infty \alpha c(\tau,x=0)d\tau = \frac{N_1}{2} - \frac{N_2}{2}$$

Thus, $N_2/2$ being the total evaporated activity the initial amount of activity must be equal to

$$N_{\text{init}} = N_1 + \frac{N_2}{2} \tag{5}$$

The final solution utilizing (4) and (5) is given by

$$c(\tau,x) = \frac{N_{\text{init}}(v-2\alpha)}{(3v-2\alpha)S\sqrt{\pi D\tau}}[\exp(-\frac{(x-v\tau)^2}{4D\tau}) + \frac{v+2\alpha}{v-2\alpha}\exp(-\frac{(x+v\tau)^2}{4D\tau})] \tag{6}$$

Depending on the value of α this solution is able to describe different cases, such as infiltration, impermeable boundary and evaporation. For longer time radioacti-

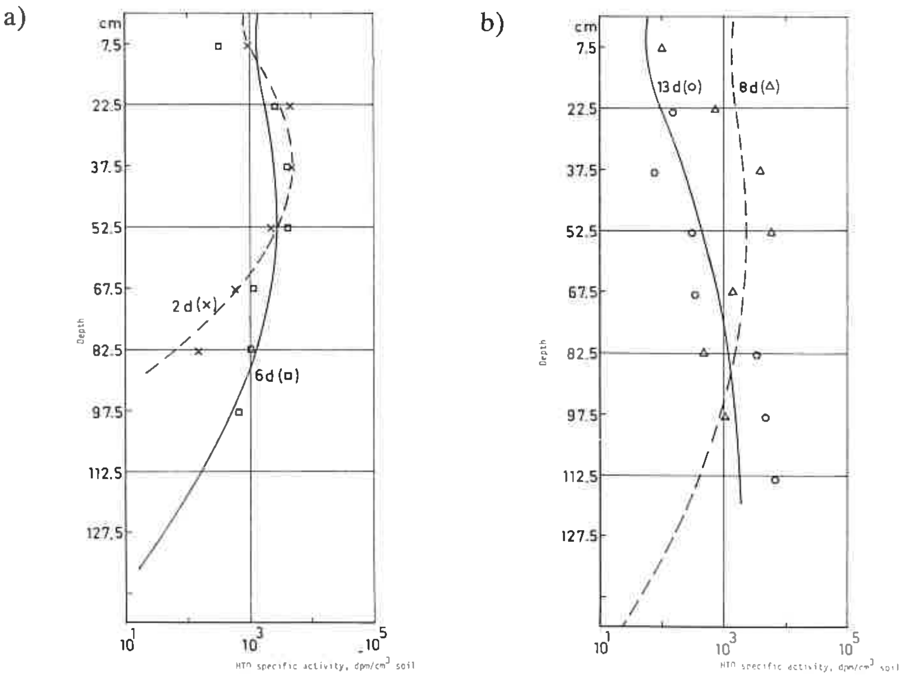


Fig. 10. a, b. Comparison of theoretical and experimental tritium distributions in gravel. $D=7.14\cdot10^{-4}$ cm^2/s; $v=15.5$ cm/d (2 d); $v=8.34$ cm/d (6 d); $v=6.85$ cm/d (8 d) and $v=9.22$ cm/d (13 d).

ve decay of initial inventory must be added. Solution (6) was found to be the best fit in comparison with other solutions. Computed results and their comparison with the measurements (plot 5, gravel) are drawn in Figs. 10-11. The results show reasonable agreement especially when the tritium moves mainly in the unsaturated zone of the soil (Fig. 10a).

Values of α and v were determined from experiments, and K_d was taken zero. Value of D was taken according to Matthess et al. (1978) and checked against the current measurements.

According to measured values most of activity was lost before the 2nd day with evaporation. This enabled us to use the solution for the infinite medium with instantaneous initial conditions but reduced initial inventory for the subsurface flow (expressed as the cumulative tritium flux at the lysimeter bottom ($x = L$)).

$$S\int_0^T F(\tau, x=L)\,d\tau = \int_0^T \frac{N_1(L+v\tau)}{4\sqrt{\pi D\tau^3}} \exp\left(-\frac{(L-v\tau)^2}{4D\tau}\right)d\tau$$

These results are plotted in Fig. 11.

Comparison of theoretical and experimental results show that the agreement is

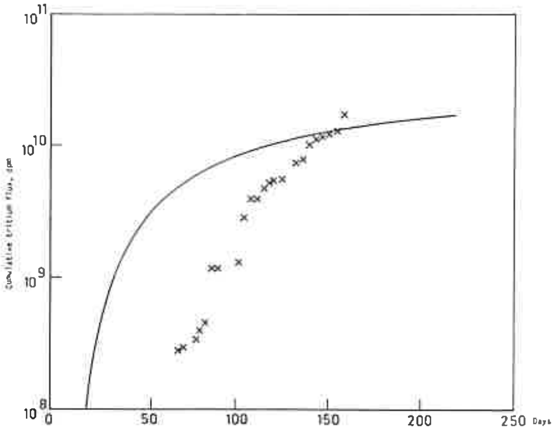


Fig. 11. Experimental (x) and theoretical cumulative tritium flux at the lysimeter bottom in gravel.

quite good while the bulk of activity is moving in the unsaturated zone. When it reaches the ground-water table the conditions for the movement change and the agreement becomes poorer. This is due mainly to the slowing down of the movement of the impulse at the ground-water table (~ 60 cm) owing to the fact that the gravitation and capillary forces are no longer seriously governing the process. It results in higher values of the experimental points at depths of about 50-60 cm, and lower values nearer the soil surface.

This change in the velocity (v) cannot be described by the theoretical solution because it was taken as a constant during the integrations of (1).

The retention phenomena in the saturated zone mentioned above can be observed also in Fig. 11. It is clearly seen that during the experiments the activity appeared at the lysimeter bottom considerably later than predicted.

Conclusions

Our experiments show that the movement of tritiated water is very different in the two types of soil (gravel and fine sand). The velocity of movement was much greater in both of these soils than in a field experiment in Ruotsinkylä (fine sand ~ 40%, clay ~ 10%, silt ~ 50%, with plants (Salonen and Miettinen 1976)). The tritium labelling gives true information about the movement of the labelled water package.

From the results in the three plots it can be concluded that the type of soil has a greater effect on the movement of tritiated water than the amount of precipitation. Under the same amount of rain + irrigated water the total activity of tritium in gravel (No. 5) was under 75 cm 13 days after labelling and after 150 days the gravel contained practically no activity any more; the fine sand No. 3 contained the maximal tritium activity between 15-30 cm after 34 days and 1 year after labelling the total amount of tritium was in this plot between 60-135 cm. The velocity of infiltration was 16-6 cm per day in gravel and 0.5 cm per day in fine sand.

The amount of precipitation has less effect on the movement of tritiated water than the type of soil. Additionally, phenomena connected with soil frost and evapotranspiration influence the results of the experiments. In the beginning the velocity of infiltration in fine sand increased from 0.5 cm to 2.1 cm per day by two times greater irrigation. This amount of »precipitation« used in our experiment (30 mm and 60 mm, respectively) is very unusual in Finland. In Hungary, however, 30 mm is not uncommon.

It means that under normal precipitation conditions the activity would stay in the upper layer for a long time and the plants could continuously incorporate the tritium from the fine sand. Uptake of the labelled water would be possible for plants with long roots even after 1 year.

The velocity of infiltration measured in Finland can be compared with that measured in Denmark where the advance of tritium from the environment into glacifluvial sand deposits has been measured (Andersen and Sevel 1974). There the velocity was about 1.3 cm per day. The activity was followed for several years in a sand layer of about 25 m.

The loss of the activity (~ 40% for gravel and ~ 30% for fine sand calculated from the total activity 1 day after labelling) was high due to evaporation. The total activity is deviated by an other type of error not considering that of the sampling procedure (~ 25%): the error of the labelling. The sprinkling method we used does not give uniform activity at the beginning of the experiment and this gives the greatest error of the calculation. Another problem is encountered when sampling under the ground-water table. The activity of the first sample is correct under the ground-water table but in the case of further course; the active water may flow into the hole from the most active layer above and disturb the measurement.

The conclusion is that labelling should be performed in an evaporation-free period of the day, e.g. late afternoon in spring or night in summer. Many samples should be taken after labelling at 0-15 cm to survey the uniformity of the labelling. If it is necessary to know the activity of the soil water below the ground-water table another sampling method should be used for it (e.g. to take the whole soil core under ground-water table with a plastic tube and then cut it into 15 cm parts).

The best fit diffusion coefficient found in gravel ($7.14 \cdot 10^{-4}$ cm^2/s) was with one order of magnitude greater than self-diffusion coefficient of water (Erdey-Grúz, Inzelt and Fodor-Csányi 1973). The theoretical solution described in this presentation is suitable to predict the movement of tritium impulses in the unsaturated zone of the soil if this zone is deep enough and the impulse is moving regularly, but in cases when the ground-water table is high, the numerical solution of the fundamental equations and boundary conditions is needed to obtain a wide range solution.

Further experiments should be performed in clay lysimeters as well as in fine sand and clay plots covered with grass. Not only the tritium activity of soil profiles but that of the grass, too, should be analyzed.

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