

STUDY OF TRANSPORT IN UNSATURATED SANDS
USING RADIOACTIVE TRACERS

W.F. Merritt
Biology and Health Physics Division
Atomic Energy of Canada Limited
Chalk River Nuclear Laboratories
CHALK RIVER, Ontario
KOJ 1J0

J.F. Pickens
Hydrology Research Division
Inland Waters Directorate
Environment Canada
OTTAWA, Ontario
K1A OE7

G.B. Allison
Commonwealth Scientific
and Industrial Research Organization
Division of Soils
GLEN OSMOND, South Australia 5064

STUDY OF TRANSPORT IN UNSATURATED SANDS
USING RADIOACTIVE TRACERS

W.F. Merritt
Biology and Health Physics Division
Atomic Energy of Canada Limited
Chalk River Nuclear Laboratories
CHALK RIVER, Ontario

J.F. Pickens
Hydrology Research Division
Inland Waters Directorate
Environment Canada
Ottawa, Ontario

G.B. Allison
Commonwealth Scientific
and Industrial Research Organization
Division of Soils
Glen Osmond
South Australia 5064

INTRODUCTION

A laboratory experiment was conducted to investigate the mixing that occurs as a series of labelled pulses of water are transported by gravity drainage down through a sand filled column having a water table imposed at the bottom. It also demonstrated the utility of gamma-ray emitting radioactive tracers in studying transport in unsaturated or saturated porous media. The motivation for pursuing this topic was developed from observing that the content of oxygen-18, deuterium and tritium in rainwater shows marked temporal variations whereas their concentrations below the water table in shallow ground water flow systems are generally found to show much less variation.

EXPERIMENTAL METHODS

The sand used in this study was taken from the east side of the Perch Lake basin at the Chalk River Nuclear Laboratories (CRNL) of Atomic Energy of Canada Limited. This sand was air-dried and then packed into a 10.1 cm (inside diameter) by 200 cm long polyvinyl chloride (PVC) column. The lower end of the column had a lucite end plate sloping slightly to a central collection tube. Water from this collection tube flowed to tubes in a fraction collector. The base of the column of sand was maintained at zero atmospheric pressure throughout the experiment. This arrangement is physically identical to the existence of a water table at the column base. The input pulses of water and tracer were added with a pump and allowed to drip onto a layer of filter paper on top of the sand column. This produced a relatively uniform infiltration rate over the top of the sand surface.

The uniformity in the initial bulk density was checked at 5 cm vertical intervals along the length of the column by transmission gauging using the 103 keV gamma rays from a 200 μ Ci source of ^{153}Gd (half-life of 242 days). The radiation was detected by a 50 mm by 12 mm thick NaI (Tl) crystal coupled to a 50 mm multiplier phototube. The detector was connected to a CRNL portable count-rate meter AEP 2160 and a CRNL Health Physics portable scaler AEP 5226. The movable source-detector apparatus consists of a bracket with the gamma source located on one side of the column and a collimator and detector located on the opposite side. The detector is shielded on the sides by lead.

An experiment was conducted to study the transport of pulses of water through the column of unsaturated sand using radioactive tracers. The pulses consisted alternately of tracer-free water and water spiked with ^{131}I and tritium. The pulses had a volume of 400 ml and were equivalent to a 5 cm layer of fluid over the column area. Each pulse was applied over a one-hour period and there was a 36 hour interval between the beginning of each pulse. By the beginning of application of a pulse, the effect of the previous moisture pulse had been transmitted through to the lower portion of the column. The duration of the experiment was 18 days during which a total of 12 pulses were added to the column. Samples were collected in the fraction collector over 0.5 hour intervals. These samples were weighed to obtain outflow rates at the base of the column. One mL samples of the column effluent samples were counted for ^{131}I on a Packard gamma counter with a 75 mm by 75 mm well-type NaI crystal. All sample activities were corrected for radioactive decay to the time at which the tracer test was started. One mL samples were also analysed for tritium using a liquid scintillation counter.

A gamma-ray transmission gauging apparatus was used to obtain moisture content profiles at various times. Calibration was accomplished by comparison of counting rates for the column filled with air-dried sand and with saturated sand. The ^{153}Gd source was removed whenever the column was scanned to detect gamma rays from tracer (^{131}I) water within the sand column.

RESULTS AND DISCUSSION

The Chalk River sand contained 93% fine sand, 7% silt and 0% clay (Unified Classification System). The approximate bulk density of the sample packed in the column was $1.66 \text{ g}\cdot\text{cm}^{-3}$ with a total porosity of 0.38. The results of the gamma-ray transmission gauging indicated that the column of sand could be considered uniform.

The outflow rate at the base of the column was measured as a function of time and outflow volume during the 36 hour periods. It varied cyclically from about 0.10 to $0.28 \text{ mL}\cdot\text{min}^{-1}$ reaching a peak about 14 hours after the beginning of the addition of a pulse at the top of the column. This variation in outflow rates is small when compared to the much more extreme flow conditions imposed at the top of the column. There pore water velocities vary by three orders of magnitude from the period of infiltration during a pulse application to the time of the end of drainage. This demonstrates the capability of the unsaturated zone to greatly dampen hydraulic conditions during drainage and redistribution of infiltrated water.

The results obtained from scanning the column with the gamma-ray detector apparatus immediately before addition of the sixth pulse are shown in Figure 1. This illustrates the displacement of the pulses at the end of drainage before the next pulse is added to the top of the column. The compression in length as they enter regions of higher moisture content in the lower portion of the column is clearly illustrated. The relative length of each slug that has been applied is shown on the figure. The regions along the column which have a higher counting rate for gamma-rays are denoted with an "I" and correspond to the location of a pulse of water that was spiked with the tracer ^{131}I whereas regions of lower counting rate correspond to the location of a pulse of water that was tracer-free. As a result of mixing as the pulses are transported down through the column there appears to be no region at depth which is completely void of tracer. The gamma scan results indicate relatively little mixing of the pulses until they reach the lower portion of the column. In this lower part of the column the pulses are contracted greatly in length due to increasing moisture content and also they are transported much more slowly. Thus diffusional redistribution from one pulse to another is enhanced leading to concentration averaging.

The results obtained from scanning the column during moisture redistribution after addition of pulse #6 are shown in Figure 2. The time of -1 h corresponds to 1 hour before pulse #6 of tracer-free water was added to the top of the column and 1.3 h corresponds to just after the pulse addition. The moisture content (θ) distribution is shown in this figure by the dashed line and the scanning counting results by the solid line. At time -1 h a tracer-spiked pulse was

located from 0 to 70 cm depth. After the pulse addition of tracer-free water, the tracer-spiked pulse is located from 25 cm to 70 cm depth and has a much higher counting rate especially in the uppermost portion of this zone. This contraction of pulses followed by expansion after the moisture front passes continues along the length of the column. This figure also shows that when a pulse of water is added to an unsaturated porous medium, this pulse penetrates only to shallow depths displacing moisture ahead of it.

The relative magnitudes of the mechanical dispersion coefficient and the molecular diffusion coefficient vary significantly both spatially and temporally during infiltration and redistribution of the input pulses. In the upper part of the column, the pore water velocities are relatively high during and immediately after the infiltration of the input pulses and hence mechanical dispersion dominates over molecular diffusion. Once the input pulse has drained to greater depth, pore water velocities decrease greatly in the upper part of the column and hence molecular diffusion becomes dominant. Pore water velocities in the lower part of the column do not exhibit the large variation in magnitude that is present in the upper part. It is expected that diffusion is important in this lower region because velocities are relatively low, the moisture content is high and the individual pulses are contracted in length.

The results of counting 1 mL samples from the column effluent samples for ^{131}I and tritium are shown in Figure 3. The relative concentration plotted versus the column effluent volume produces a sinusoidal curve after the first spiked water has been eluted. Each 400 mL of effluent is equivalent in volume to one input pulse. Initial breakthrough of the first tracer-spiked pulse occurred after 5.5 pulse volumes. The deviation of this breakthrough curve from a square wave is a measure of the amount of mixing or dispersion that has taken place. The peaks and troughs of the tritium curve exhibit a gradual dampening to the right. There was a 4% difference in travel rates between tritium and ^{131}I . For the conditions imposed in this experiment it appears that significant concentration variations can be transmitted through the unsaturated zone to the water table.

CONCLUSIONS

Pulses of water of different concentrations existing in a soil profile will expand and contract greatly in length depending on the location of the moisture front. The contracted size of individual pulses as they enter the saturated zone leads to enhanced diffusional redistribution. The contracted size of pulses after entry to the saturated zone necessitates water sampling at a very fine scale. Otherwise, an averaged sample of different ground waters will result.

The relative magnitudes of the mechanical dispersion and molecular diffusion coefficients vary temporally and spatially during infiltration and redistribution of the input pulse.

There was a 4% difference in travel rates between tritium and ^{131}I . This effect was illustrated clearly for a 2 m column but would have been difficult to define in shorter columns. This also raises the common question as to what constitutes an ideal tracer.

The use of gamma-emitting tracers such as ^{131}I has been demonstrated to be useful in studying transport in soil columns. It could also be used in field studies of infiltration and mixing in the unsaturated zone. There, a collimated detector could be lowered inside a dry access tube to detect the tracer within the soil water.

COLUMN GAMMA-RAY SCANNING

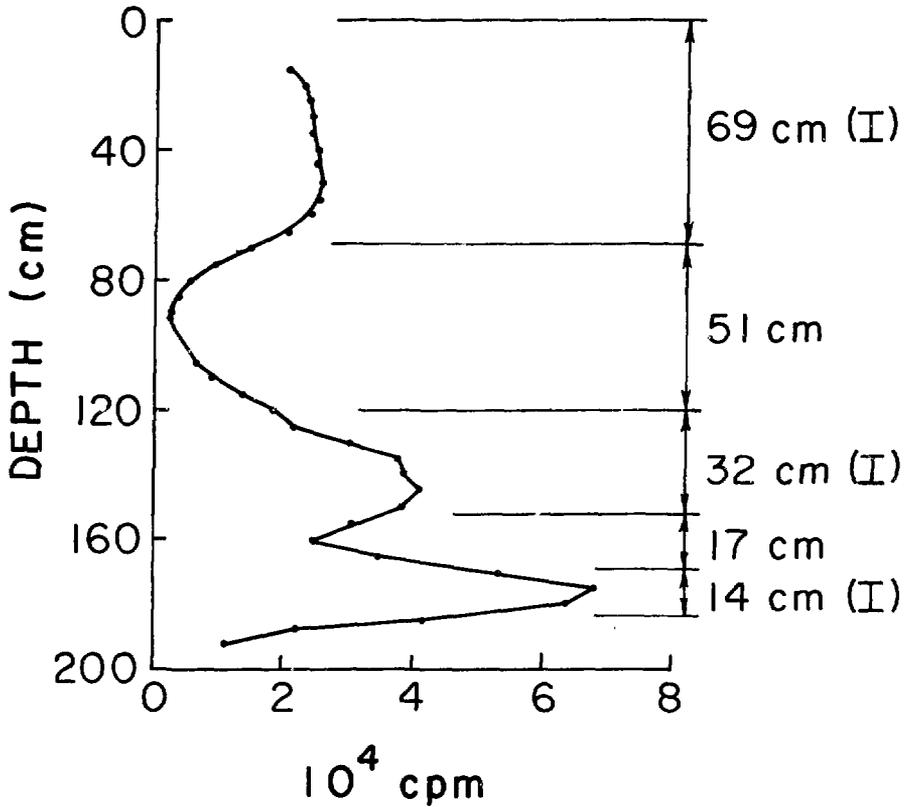


Figure 1. Gamma scan results along column immediately before addition of the sixth pulse.

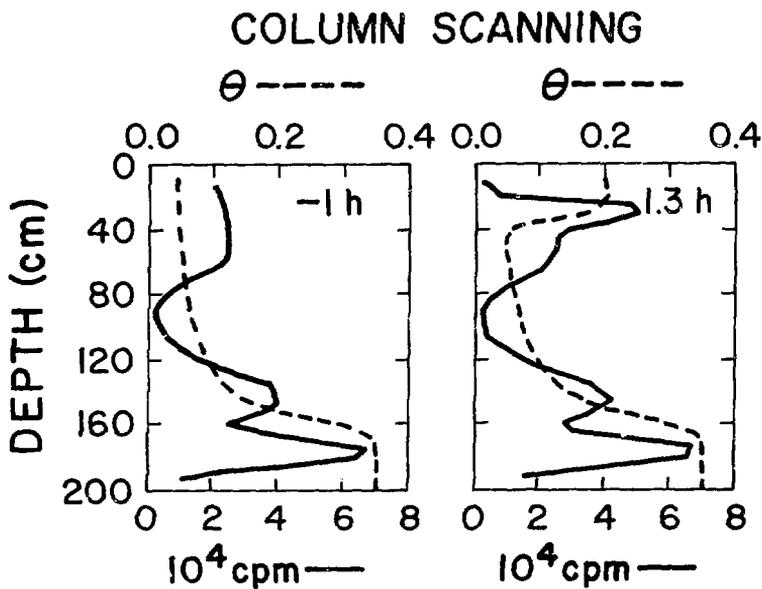


Figure 2. Gamma scan results along column during moisture redistribution after addition of the sixth pulse.

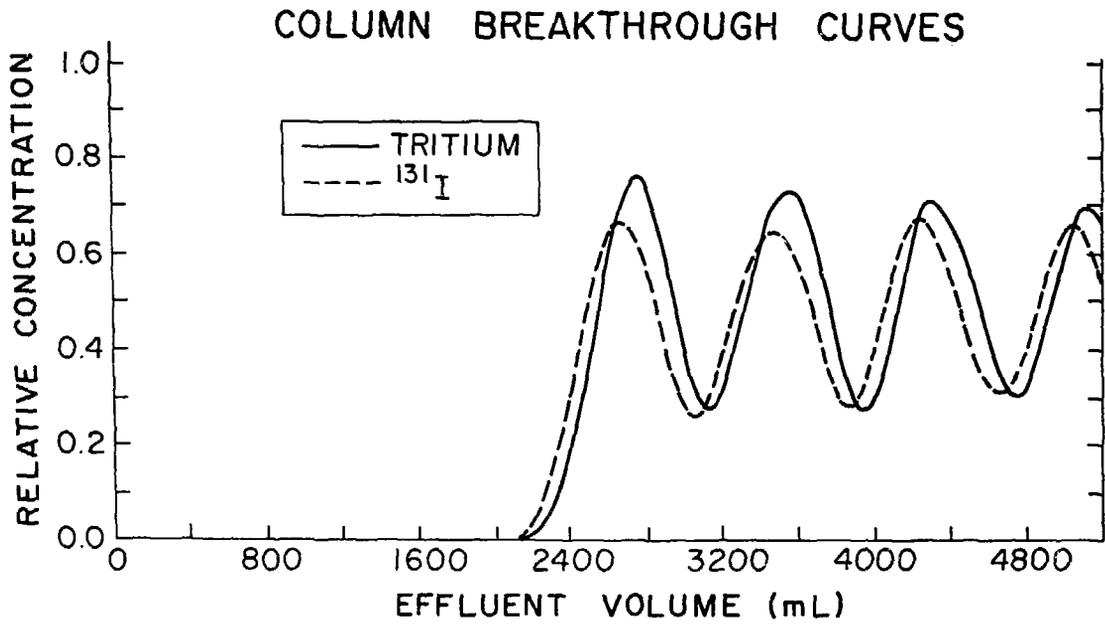


Figure 3. Column breakthrough curves for tritium and ¹³¹I tracers.