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To: U.S. Atomic Energy Commission
Division of Biology and Medicine

Title: The Mixing Process in Soils of Solutions of Different Densities
and Viscosities

Institution and Department: Cooperative research between Agricultural
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PROGRESS REPORT

Scope:

In this project we are studying, by laboratory experiments, stability and mixing characteristics of flow of solutions that displace other solution in soil. Density and viscosity gradients are known to be main factors that influence the stability in the mixing process. However, most of the experimental studies on stability in miscible displacements have been carried out in laboratories of petroleum engineering. The density and viscosity ratios, permeabilities, and flow velocities in these experiments are much larger than those expected in agricultural soils. In the third year's work (the subject of this report), we carried out experiments on stability using soil as a porous medium. In our work we used CaCl_2 solutions of different concentrations and used soil columns of different length to study the mixing process. Miscible displacements were made by introducing the first solution into the soil column to displace the second solution under the steady state flow condition. Either the displacing or displaced solution was labelled with Cl^{36} to obtain breakthrough curves which show the mixing characteristics. The tracer concentration of outflow from the column was continuously recorded by a liquid scintillation flow system. The recorded curves revealed clearly instability of the flow under unstable conditions at 2.2 cm/hr flow velocity. The instability was observed in columns 10, 20 and 30 cm long with density and viscosity differences of 1.75×10^{-3} g. per ml and 2.6×10^{-3} centipoises, respectively. Apparent dispersion coefficients (units, cm^2/hr) calculated from the breakthrough curve were 0.224, 0.258 and 0.306 for 10, 20, and 30 cm column, in order, showing that the apparent coefficient increases with the column length. (The theory used for

Scope: continued

calculating dispersion coefficients is based on semi-infinitely long columns.) The longest column used was not long enough to stabilize the unstable flow. The unstable flow was considered to be due to the high permeability of the porous medium and the large flow velocity, so that the fluids left the columns before they could thoroughly mix and be stable.

Experimental:

Three columns of soil were used in this study. The columns were 10, 20, and 30 cm long, having the same radius of 3.8 cm.

Samples of air-dry soil were packed into the columns so that the same bulk density could be obtained. The gamma ray density meter was used to help obtain uniform density. The soil was Hagener sandy soil with a textural composition of sand 92%, silt 2% and clay 6%. The bulk density was $1.45 \pm 0.03 \text{ g/cm}^3$.

To ensure complete water saturation, entrapped air in the soil column was flushed out by introducing CO_2 gas for one hour before water was allowed to enter the column. Water saturation was made with 0.01 N CaSO_4 solution in the vertically upward flow direction. The soil columns were treated with 0.5 N CaCl_2 solution to saturate the soil particles with Ca^{++} . A day after this treatment, the 0.5 N CaCl_2 solution was displaced by 0.01 N CaCl_2 solution until no further change in Cl^- concentration in the leachate was observed. When a steady-state flow condition was established with 0.01 N CaCl_2 solution, the column was ready for the miscible displacement study for saturated flow. The saturated hydraulic conductivity was determined with 0.01 N CaCl_2 after the steady-state condition was established. The

Experimental: continued

The hydraulic conductivities were 30.7, 32.9, and 32.7 cm/hr for the 10, 20, and 30 cm long column, respectively. These nearly equal hydraulic conductivity values show that the three columns were packed essentially the same.

The miscible displacements were made for both directions, vertically upwards and downwards. The experiments were performed in the constant temperature room controlled at $20 \pm 0.5^\circ \text{C}$. All solutions were either 0.01 N, 0.05 N, or 0.10 N CaCl_2 solution. Densities and viscosities of the solutions were measured at 20.1°C . The measured densities of these solutions were 0.99875, 1.00050, and 1.00273 g/cm^3 with the corresponding measured viscosities of 1.0029, 1.0055, and 1.0153 centipoises. Either displacing or displaced solution was labelled with Cl^{36} . The activity of the solution was 1.6 microcuries per liter.

As in the second year's experiments, a liquid scintillation flow system (Nuclear Chicago Model #4526) was used to measure and record continuously the tracer concentration of the effluent from the column. The system consists of a detector, a beta-ray-analyzer, a ratemeter, and a chart recorder. The soil column was connected with the flow cell in the detector chamber of the system. The effluents from the column were permitted to pass the flow cell. The flow rate was controlled by means of a constant volume pump placed between the column and the flow cell. As the effluent passes the flow cell, the tracer is detected and the concentration of the tracer is recorded on the chart recorder. The recorded curve shows the continuous change in concentration of tracer. A small flow cell was used to obtain better volume resolution so that instability irregularities

Experimental: continued

following each other closely could be observed. The fluid volume of the cell is 1.15 ml, the ratemeter time constant setting is 50 seconds. Thus, the recorded activity is an average over 50 seconds. Chart speed is 6 in/hr. The 6 inch interval of the chart paper is divided into 8 divisions. At a flow rate of 40 ml per hour, each division is equivalent to 5 ml of effluent.

A difficulty was encountered in using the anthracene flow cell this year. In some of our columns we used phenol to prevent bacterial action. This phenol got into the flow cell, fouled it and changed its calibration. We discontinued the use of phenol. Tests showed that we could get by without a bactericide for the times of our experiments. In the meantime Nuclear Chicago has supplied a glass flow cell that we should be able to use with phenol if necessary.

Breakthrough curves were drawn from the recorded curve by plotting concentration versus pore volume. The concentration is presented as a ratio of height of the recorded curve at a given time to a maximum height of the curve. The maximum height corresponds to the original concentration of solution entering the column. For the maximum height, an average was taken over a one hour period after displacement was completed. Pore volume is calculated from the following relation:

$$\text{pore volume} = \frac{V}{L} t, \quad V = \frac{\bar{V}}{P}$$

where V is the flow velocity in cm/hr, L is the column length in cm, t is the time in hours and t is the elapsed time after the displacing solution is introduced to the column, $\bar{V}$ is the Darcy velocity in cm/hr and P is the porosity.

Results and Discussion:

The results are shown in seven figures that will be discussed in order. The first figure, Fig. 1, indicates how well the flow cell system works.

Enter Fig. 1

A schematic of the system is shown at the top of the figure. The bottom shows two of our example curves recorded by the system. These curves were obtained with a 10 cm long soil column, where the miscible displacement was for a 0.10 N solution displacing a 0.01 N solution in two vertical flow directions, "downward flow" and "upward flow", at almost identical flow velocities, 2.2 cm/hr. The curves show change in concentration of tracer in the effluent with time. Time scale on the abscissa moves from the right to the left. On the ordinate axis, concentration of tracer is shown with an arbitrary unit. In all of our experiments, where displacement is complete, the concentration scale reaches about 90% of total scale, which is equivalent to about 2700 cpm. The upper reading of the concentration scale depends on the ratemeter setting as well as the radioactivity of the initial solution and the efficiency of the flow cell. Breakthrough times are shown by arrows at the right hand side of the curves of Fig. 1. Some further details may be given about the curves of Fig. 1 to show what data is associated with the charts. The top curve of the figure is for tagged 0.10 N CaCl_2 solution displacing nontagged 0.01 N CaCl_2 solution, in the vertically downward flow direction. The bottom curve is for the reverse flow direction. For the downward flow, the displacing solution (0.10 N) appears in the effluent before 3 hours, while for the upward flow the displacing solution is detected after 3 hours. Since the two flow velocities

Results and Discussion: continued

are very slightly different, the difference in breakthrough time is caused mainly by the difference in concentrations associated with the flow direction, not velocity. Some fluctuations within small time ranges are seen on both curves. These minor fluctuations are due to the random nature of radioactivity. The downward flow curve shows an intermittent change in slope, unlike a gradual change for the upward flow curves. The characteristic intermittent shapes of the downward flow are considered to be an indication of unstable flow. The breakthrough curves for the 10 cm long column of Fig. 1 will be presented later. However, it should be pointed out that some information on the mixing process can be obtained directly from a recorded curve before a complete breakthrough curve is drawn. This continuously recording system eliminates uncertainty in what happens between data points obtained when one analyzes increments from an outflow as in other analytical procedures.

Fig. 2 presents breakthrough curves obtained with a 30 cm long column in the vertically downward flow direction. Displacing and displaced solutions are the same except one is tagged with Cl^{36} and another is not. There is

Enter Fig. 2

actually no density or viscosity differences between two solutions. When displacement with the tagged solution was completed, the non-tagged solution was introduced to displace the tagged solution. An open circle curve is the breakthrough curve for the tagged displacing solution, and the solid circle curve is for the non-tagged displacing solution. Two

Results and Discussion: continued

curves lie on each other, showing no difference, regardless which solution is tagged. Therefore, for the following runs, it may be assumed that Cl^{36} in the low concentration used does not influence the flow characteristics and gives them correctly. In the figure, the solid line is a theoretically calculated breakthrough curve. The behavior of the tracer is fairly well predicted by the calculated curve. The good agreement between the experimental and the calculated curves may result from the fact that sandy soil was used. We suppose that in soils with much clay, absorption phenomena would rule with the simple theory used here. Our sand had 6% clay. It was not a synthetic washed sand.

Fig. 3 shows two breakthrough curves obtained with a 20 cm long column. Displacements were made between 0.10 N and 0.01 N solutions. The flow directions are vertically upward. The solid circles are for an 0.10 N

Enter Fig. 3

solution displacing an 0.01 N solution (stable situation), and the open circles are for the 0.01 N solution displacing the 0.10 N solution (unstable situation). When the denser solution is above the lighter solution and is displaced by the lighter solution, the breakthrough curve is irregular in shape and has an overall less steep slope. One notes discontinuities in the derivative of the open circle (unstable) curve of the figure.

Aside from the overall slope differences and discontinuities in the slope of the "unstable" curve, there is another main difference seen between the stable and unstable flow. For the unstable flow the tracer of the displacing solution appeared in the effluent when 0.6 pore volumes entered the column. For the stable flow, the tracer of the displacing solution was detected when 0.7 pore volumes entered the column. On the other hand, the displacement

Results and Discussion: continued

of the stable flow was completed earlier than that of the unstable flow. The earlier breakthrough and the prolonged displacement are an indication of a more rapid, an earlier and later mixing caused by the unstable flow.

Fig. 4 shows breakthrough curves obtained with a 30 cm long column. Except for column length the conditions for the figure are as for the preceeding figure. The breakthrough curves in the figure show differences

Enter Fig. 4

between the stable flow and the unstable flow. The tracer of the input solution appeared in the effluent earlier under the unstable conditions (open circles) than under the stable conditions (solid circles), and it required more input solution for the unstable flow to complete the displacement. These earlier breakthrough and delayed displacement of the unstable flow were already observed with the 20 cm long column. However, there is some differences between the unstable breakthrough curves of the 20 cm and 30 cm long columns. Under the same unstable conditions, the 30 cm long column required about 1.3 pore volumes of the displacing solution to complete displacement while the 20 cm long column needed almost 1.5 pore volumes. Thus, the curve of the 30 cm long column is steeper than that of the 20 cm long column.

Figs. 5, 6, and 7 are all for downward miscible displacement. The three figures are for the same conditions except for the column length:

Enter Figs. 5, 6, and 7

Fig. 5 is for 10 cm length, Fig. 6 for 20 and Fig. 7 for 30 cm. On each figure, breakthrough curves for 0.05 N or 0.10 N solution displacing a

Results and Discussion: continued

common 0.01 N solution are compared with the third curve for 0.10 N solution displacing itself. For each column length, the breakthrough curve (solid circle) of the displacement, between the same concentrations of 0.10 N, has a regular shape. This curve is for stable flow. On the other hand, the curve for 0.05 N solution displacing 0.01 N solution (crosses), and the curve for 0.10 N solution displacing 0.01 N solution (open circles), each has an irregular shape. The irregular shape shows unstable flow. For the unstable situation the curves show that the heavier solution always broke through first and took a longer time to complete the displacement, that is, to reach a unit value of relative concentration. In general, for the 3 columns of different length, the breakthrough pore volumes are about the same for all three columns, but the pore volumes required to complete the displacement are different. The longer column required fewer pore volumes to complete the displacement.

The apparent dispersion coefficients for the 0.10 N CaCl_2 solution displacing itself for the three column lengths of Figs. 5, 6, and 7 were computed on the assumption the columns were semi-infinitely long. For Fig. 5, the 10 cm column, the dispersion coefficient was $0.224 \text{ cm}^2/\text{hr}$; for the 20 cm long column, 0.258 and the 30 cm column, $0.306 \text{ cm}^2/\text{hr}$. The fact that the values are not equal shows that 10, 20 and 30 cm long columns may not be considered of infinite length except as rough approximations. (No theory is available for calculating a dispersion coefficient when one concentration displaces another in either finite or semi-infinite flow. Most roots and most chemical process as from insecticides, etc. occur in a layer of top soil less than 30 cm deep.)

Results and Discussion: continued

Our previous figures (Fig. 3 through 7) have shown that in our experiments, the flow was unstable regardless of the flow direction when the heavier solution was above the lighter one. The density and viscosity differences between 0.01 N and 0.05 N CaCl_2 solutions were $1.7 \times 10^{-3} \text{ g/cm}^3$ and 2.6×10^{-3} centipoises, respectively. Instability was observed in 10, 20 and 30 cm long columns at the flow velocity 2.2 cm/hr. The column length used did not result in stabilized flow. It appears that a slower velocity or a longer column will be needed before the flow will become stable, if one wants it stable. The column lengths 10, 20 and 30 cm were used for convenience and are also used because, as has been indicated, most crop plant roots occur in a depth of less than 30 cm of soil.

Using our third year's data we were able to see whether variables in our experiments (densities, viscosities of the solutions, permeability, flow velocity, self dispersion coefficients) gave an unstable flow parameter in agreement with Dagan's theory (report C00-1269-8). The stability parameter that we calculate from our data is $\lambda = 0.45$. Dagan's theory says this value should give quasi-stable flow. Our value 0.45 of λ is subject to revision as we computed it on the assumption that the longitudinal and transverse dispersion coefficients are the same. The transverse dispersion coefficient will receive further study.

A paper was presented "Density and viscosity effect in the soil water mixing process" by Sun-Ho Yoo and Don Kirkham, American Society of Agronomy Meeting, Detroit, Michigan, November 13, 1969.

Work on a publication has been completed and it is now in print: Fritton, D.D. "Resolving time, mass absorption coefficient and water content with gamma-ray attenuation". Soil Sci. Soc. of Amer. Proc. 33:651-655, 1969. When reprints arrive, one copy will be sent to AEC.

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Typical dynamic flow counting system illustrating the basic detection, analyzing, and recording components.

From Nuclear Chicago Manual

Fig. 1

30-cm COLUMN, DOWNWARD

Fig. 2

20-cm COLUMN, UPWARD

Fig. 3

30-cm COLUMN, UPWARD

Fig. 4

Fig. 5

20-cm COLUMN, DOWNWARD

30-cm COLUMN, DOWNWARD

Fig. 7