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SOME PHYSICAL PROCESSES CAUSING MOVEMENT OF IONS AND OTHER MATTER THROUGH SOIL*

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Water is important as a mover of chemicals in, or into soil because chemicals, unless volatile, will seldom move without water moving.

I will consider three processes of water movement in soil which will result in movement of chemicals. These processes are: (a) miscible displacement, (b) diffusion and (c) leaching. These are processes which I and my students are actively investigating. We will first consider miscible displacement, and do so in some detail, as it is a new and fascinating subject in soil science. In conclusion, a short section on movement of pesticides, prepared by a colleague, will be presented.

MISCIBLE DISPLACEMENT

Water, either with or without chemicals in solution, can move into the soil, and can mix with chemicals there. When the water mixes with

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the chemicals which are in the soil, some of these chemicals will move with the water, at a slower or faster rate, depending upon the size of the pores at the point where the water encounters the chemicals, and depending upon other factors. This process of mixing with, and displacing chemicals is called miscible displacement.

Piston flow -- The most complete type of miscible displacement is, in fact, not a mixing process at all, but is a 100% displacement process called piston flow. A schematic drawing illustrating piston flow is presented in Fig. 1. In the upper part of the figure there is a tube with a

Insert Fig. 1. Piston flow in a tube.

piston at the left-hand end and a small outlet tube at the other end. Over the first portion of the inside of the tube there is sodium chloride or ordinary salt solution. Initially we may think of the sodium chloride as being separated by a membrane from some water. The water is at the right of the sodium chloride solution. But we can pull out the membrane and then push the piston to the right. If we push the piston fairly rapidly so that there will not be time for molecular diffusion to occur across the boundary between the salt and the water, the water will be moved to the right without any mixing at the interface, as is seen at the middle part of the figure. Only water will come out until the salt front reaches the end of the tube. Then, only salt solution will come out, indicated at the bottom of the figure. In piston flow the salt solution must proceed with the same velocity everywhere within the tube.

Miscible displacement in a capillary tube -- In Fig. 2 we show how salt solution moves through a capillary tube if we assume there is no molecular diffusion. In the upper part of the figure notice that as the

Insert Fig. 2. Capillary flow in a tube assuming no molecular diffusion. (Nielsen and Biggar, 20.)

salt (sodium chloride) is pushed into the tube from the left, the salt moves more easily along the axis of the tube than it does along the walls. This is because the fluid next to the wall of the capillary tends to stick there. In fact, when we derive Poiseuille's law for capillary flow, we assume that the velocity of the fluid next to the wall of the capillary tube is zero.

In the lower part of the figure the salt front has advanced much farther along the axis; it will not be long before sodium chloride will move out of the capillary tube and be found in the outflow water. Then, as additional sodium chloride is moved through the tube, the concentration of the salt in the droplets of the outflowing liquid will increase. In capillary flow it is not a case of all water, and then all salt coming out, as in piston flow. In capillary flow, the smaller the diameter of the tube, and the longer the capillaries, the greater is the mixing.

Flow in saturated soil -- In Fig. 3 we see how the input solution will move through water-saturated soil. Since there are both large

Insert Fig. 3. Flow in saturated soil.

pores and small pores in the soil, we will have a combination of piston

flow and capillary tube flow. Water in the smaller soil pores will move slower than in the larger pores if the larger pores are continuous to the outflow end. The small pores will tend to hold the original soil water. A large volume of input solution will be required before the water in these smaller pores will be replaced by the input solution and the concentration of the outflow solution becomes that of the input solution.

Flow in unsaturated soil -- Unsaturated flow is the most common type of flow found under field conditions. Unsaturated flow occurs largely by a process often called capillary diffusion. This process is a much more complex process than is saturated flow. In Fig. 4 we show the relation of soil particles, water, and air spaces in a cross section

Insert Fig. 4. Flow in unsaturated soil.

of an unsaturated core. The large pores, which conducted the salt solution easily under saturated conditions, are partly filled with air and have lost much of their conducting effectiveness. The salt solution must move through the tortuous paths presented by the smaller pores. To complicate matters further, many pores are blocked by air after a short distance. These pores are "dead-end" pores and do not permit flow to occur through them. When "dead-end" pores are present, molecular diffusion is important for complete mixing of the input solution with the soil solution.

In the process of miscible displacement I indicated that molecular diffusion (a subject we will consider in some detail later) may be a

factor. In fact, if the water of the miscible displacement process moves into, or in the soil at practically zero velocity, so that the salt front of our Fig. 1 stays in contact with the water front for a long time, some of the salt molecules may move forward and completely out of the tube, to the right, just through molecular activity, called (molecular) diffusion. I believe we are all familiar with the classical diffusion experiment where one puts copper sulfate crystals in the bottom of a column of water and observes how, over the months, the blue color will gradually rise in the column of water due to the movement of the copper sulfate molecules up into the clear liquid above.

Analyses of Miscible Displacement Data

Now the question arises, how do we analyze the data of the miscible displacement processes? Actually, in our experiments, to see how soil behaves with regard to the movement of these chemicals into the soil water system, we let the inflow solution come in at one end of a soil sample and then examine the outflow solution. As shown in Fig. 5, we plot, on the ordinate, the concentration of the tracer solution at the out-

Insert Fig. 5. Way of presenting miscible displacement data.

flow side of the tube divided by the concentration of the tracer solution at the inflow side of the tube, while on the abscissa we plot the volume of solution collected at the outflow divided by the volume of solution in the soil pores. The concentration ratio on the ordinate is denoted,

$$C_{\text{out}}/C_{\text{in}}$$

The volume ratio on the abscissa is denoted,

$$V_{\text{out}}/V_{\text{solution in pores}}$$

The volume, $V_{\text{solution in pores}}$, can change for the same soil sample because, in our work, we run the input solution into the soil when the soil is at different percentages of saturation. It is easy to send solutions into a water saturated sample, but to pass the input solution into a sample which is only partially wet and kept everywhere at the same soil-water tension is a difficult problem. The problem has been overcome (a) by developing fritted glass bead discs (by my students) and stainless steel filters (commercially) having very low impedance for fluid flow, and (b) by tilting the sample at a proper angle (see Corey et al. (5)).

Hypothetical curves for piston and capillary flow -- Now let us look at the types of flow we have discussed. Figure 6 shows the outflow curve of tracer in 100% piston flow. Here we notice that one pore volume (as

Insert Fig. 6. Outflow curve for piston flow. (Nielsen and Biggar, 20.)

the volume of the solution in pores is often called) passes out of the solution before any change in the concentration ratio is observed. Then the salt solution suddenly breaks through at the output, so that the concentration ratio $C_{\text{out}}/C_{\text{in}}$ is unity. This unity value continues as long as the input salt is passed through the tube. In the case of capillary flow, the concentration ratio versus the outflow volume is as shown in Fig. 7. There is not a complete jump in the concentration ratio from zero to

Insert Fig. 8. Outflow curve for flow in saturated soil. (Nielsen and Biggar, 20.)

unity, as for piston flow. The calculated outflow curve, assuming no diffusion, reaches unit concentration very slowly.

In actual saturated soil, the outflow curve of tracer ion (fig. 8)

Insert Fig. 8. Outflow curve for flow in saturated soil. (Nielsen and Biggar, 20.)

reaches the unit concentration ratio sooner than for capillary flow in a tube because diffusion exists. In unsaturated soil, the outflow concentration of tagged ion reaches (fig. 9) the maximum value much later

Insert Fig. 9. Outflow curve for unsaturated flow in soil.

than for saturated soil because of the increased number of discontinuous pores present in the unsaturated soil.

Experimental curves of miscible displacement -- Some actual outflow concentration curves obtained in miscible displacement experiments are available and Fig. 10 shows data obtained from an experi-

Insert Fig. 10. Outflow curve for chloride in Oakley sand. (Nielsen and Biggar, 20.)

ment using Oakley sand. Here the input, a chloride solution, passed through the sample at a rate of 0.79 cm per hour. Figure 11 shows a similar curve for a clay loam. On comparing these two figures, one can see that the concentration ratio does not build up as fast in the outflow solution from the clay soil as in the outflow solution from the sand. In Fig. 12 we see two outflow curves of tritium for Yolo loamy sand.

Insert Fig. 11. Outflow curve for chloride in Aiken clay loam. (Nielsen and Biggar, 20.)

These water contents are not much different; neither are the input velocities but, even so, the concentration curves look quite different.

Ions and materials moved by miscible displacement -- We (especially Mr. John C. Corey) are studying the movement of nitrate ions with the miscible displacement technique. Using this same technique we can also measure the displacement of other ions in the soil and of organic molecules, such as herbicides and fungicides. Certainly, nitrates and plant fertilizing ions, as they are moved by miscible displacement, are of extreme interest. To see just how these ions are moved, we are using radioactive and non-radioactive isotopes for labeling or tagging. In the case of the nitrate ion we tag the nitrogen with heavy nitrogen and then use a mass spectrometer to determine heavy nitrogen.

DIFFUSION

Our second main topic is diffusion and it is the process we mentioned in connection with miscible displacement. The diffusion process is important in periods when there is little water in the soil. In these dry periods there are two ways the plants can obtain water, with its nutrients. The roots can move either to locations where soil water and nutrients are located, or the nutrients can move in the water to the roots by the process of diffusion. There are two general types of diffusion: (1) capillary diffusion where the water moves because of a moisture

gradient; and (2) molecular diffusion where the water molecules and the salt molecules interdiffuse, as in the copper sulfate experiment we mentioned.

We distinguish between two types of molecular diffusion. The first type is called molecular self-diffusion, diffusion of molecules of the same kind into each other; the second type of molecular diffusion occurs when molecules of different kinds redistribute themselves. We shall speak here mainly of molecular self-diffusion.

When we speak of molecular self-diffusion of water, we are thinking of the way certain water molecules which have an imaginary black mark (say) on them move into the other water molecules which do not have this imaginary black mark on them. And to study how this occurs we "put the black mark on the molecules" by using a water molecule, deuterium oxide, D_2O , which is of slightly different mass than the ordinary water molecule. These heavy water molecules are sometimes denoted by H_2^2O . The heavy H_2^2O molecule weighs $2 \times 2 + 16$ or 20 units, while the ordinary water molecule, H_2^1O , weighs $2 \times 1 + 16 = 18$ units. But if there are only a few heavy water molecules, their extra weight will not appreciably interfere with a study of molecular self-diffusion by this means of tagging.

We study the self-diffusion process by taking a soil column, one half containing water tagged with heavy hydrogen molecules and the other half containing only the normal hydrogen molecules. Both halves are at the same moisture content. We allow these two halves to stand while molecular action occurs. After different periods of time, we take

sample segments from the soil columns to see how much of the heavy water has moved into the regular water. The closer the water sample is to the center, the more heavy water it will contain.

Figure 13 shows an experimental concentration function F of the

Insert Fig. 13. Molecular self-diffusion of water in Colo clay loam soil. (Kunze and Kirkham, 21.)

heavy water, plotted against distance from the center of the soil sample after 57 hours of molecular diffusion. We see in the same figure a theoretical molecular diffusion curve of the function F versus distance x , on the basis that the diffusion is a linear process; that is to say, the diffusion of the molecules is proportional to the concentration gradient of the tagged molecules. The experimental points and the theoretical curve agree well, showing that we are dealing with a diffusion process for which we have a theory.

Figure 14 is like Fig. 13 except that glass beads are the "soil". The

Insert Fig. 14. Molecular self-diffusion of water in glass beads. (Kunze and Kirkham, 21.)

figure shows an even better fit of the experimental data to the theoretical. Synthetic soils of glass beads are convenient to work with, and they give good results. Why can't true soils behave more like glass beads!

Capillary diffusion has been mentioned in connection with miscible displacement. Another aspect of our capillary diffusion work is the

development of apparatus and methodology by measuring the so-called capillary conductivity coefficient. I will not discuss this here.

LEACHING

Leaching, the rinsing of undesired salts from the soil, is an important process in land reclamation and management. The process is especially important in locations where there is little rainfall and the agriculture is carried on mainly by irrigation. When there is little or no rainfall, water tends to rise to the soil surface by capillary action carrying salt with it. The water evaporates. The salt does not evaporate, but stays in the soil. The salt concentration can build up so strongly near the surface of the soil that plants cannot grow. Fresh water is put on the surface of the field to rinse these salts out into drainage water so that the salt concentration is reduced and plants will grow again.

Figure 15 shows a leaching situation, such as may be found in Egypt

Insert Fig. 15. A leaching situation as found in Egypt. (t = thickness of surface water; $2s$ = spacing between ditches; d = distance from soil surface to gravel; h = height above gravel of water in drainage ditches; x and y = coordinates with origin 0; circled numbers are discussed in the original paper.) (Kirkham, 22.)

in the reclamation of Lake Maryut. This is a lake which has strata of calcareous sea shells about one meter below the surface of the soil. When drainage ditches are put through this land and leaching water is applied at the top, the situation is equivalent to that of this figure, which shows gravel underlying soil, with the gravel underlain by impermeable

clay. Non-saline water is applied by ponding at the surface of this soil. Some of this applied water moves out to ditches at the sides, and some moves down to the gravel and then out to the ditches. The underlying shell stratum, indicated in the figure by gravel, provides a fortunate situation for leaching. The rapidity with which Lake Maryut soil can be reclaimed amazes soil scientists. They say that the rapidity of the reclamation is a result of soil flocculation (aggregate formation) caused by the calcareous sea shells, which makes the soil more permeable and hence more leachable. This is, in part, correct, but another reason for the easy leaching is the high water conductivity of the shell layer, permitting the water to move straight down to the shells and then move against very little friction to the outflow ditches.

Figure 16 shows the theoretical stream flow pattern and lines of

Insert Fig. 16. Theoretical flow net for Fig. 15. (Kirkham 22.)

equal hydraulic head, called a flow net, for the situation of Fig. 15. We see from the nearly equal spacing of the streamlines near the surface of the soil that the amount of water that passes between unit areas near the surface is about the same over the whole distance between the ditches, so that the leaching process will proceed rapidly everywhere.

When there is water pressure from below (artesian pressure) it is very difficult to leach the soil. There is a gradual upward movement of the water, preventing the leaching water from entering the soil. There are numerous agricultural sites in the world where this upward seeping water prevents easy leaching. An example is a polder where

diked-off sea water causes upward pressure.

When a pipe is driven into artesian gravel (fig. 17), the water in

Insert Fig. 17. Surface water, for leaching, in the presence of artesian pressure. ($s + j$ = standpipe height of water above artesian gravel; for other symbols see the original paper.) (Kirkham, 23.)

this standpipe may stand 10 or even 50 feet above the soil surface showing the great artesian pressure tending to drive up water. Even if tight clay overlies the artesian gravel, there is an upward seepage process that tends to block downward movement. By the use of drain tubes in sand tank models (fig. 18) my students have demonstrated this upward

Insert Fig. 18. Laboratory test of leaching when there is artesian pressure. (Laboratory streamlines, Harding and Wood, 25; theoretical points, Kirkham, 24.)

seepage process, along with the downward seepage. If enough head is put on the surface of the soil by surface water and if the artesian pressure is very small, then we can leach the surface soil; but if the artesian pressure is great, the water midway between the drains will not go over to the drains, but will tend to rise to the surface. Thus, midway between drains there will be little or no leaching.

The existence of an impermeable layer near the surface of the soil causes a difficult leaching problem. When such an impermeable layer exists, this layer prevents the input water in the leaching process from getting into the soil easily midway between the drains. In fact, Fig. 19, which gives the inflow rate at the soil surface versus distance between

Insert Fig. 19. Water inflow rate of leaching water versus distance from drains. (d = depth of drain tube centers from surface of soil; $2r$ = diameter of drain tubes; a = spacing of drain tubes; K = hydraulic conductivity of soil; t = thickness of surface water; h = depth from surface of soil to impermeable layer; x = distance from drain tube center; Q' = volume of water per unit surface area seeping per unit time into the soil.) (Kirkham, 26.)

drains 100 feet (or about 30 meters) apart, shows that the flow rate Q over the drains is very large compared with that midway ($x = 50$ feet) between drains. There are two curves shown, one where the impermeable layer is 6 feet deep ($h = 6$ feet) and one where it is at a very great depth ($h = \text{infinity}$). When the impermeable layer is 6 feet deep (or 2 meters deep), the rate of inflow water drops off to practically zero at a distance 20 feet from the drain. So between 20 feet and 80 feet almost no water will move into the soil. But if the impermeable layer is at the very great depth, there is still appreciable water movement into the soil from the 20 foot mark to the 80 foot mark, and leaching can occur, although the leaching rate will be small compared with the rate over the tiles.

Sometimes there is a tight layer almost at the plow layer depth in the soil, so that with drainage ditches or drains a long distance apart, the movement through the soil midway between drains is practically zero. In these cases we should apply, successively, quantities of leaching water, because then a hydraulic gradient can be established to drive the water midway between the drains to the drains. What I am saying is that if the water drops down to the drain level, then the water midway

between drains, which has not dropped to the drain level, has a large difference in head between it and the drain so the water can then move out. Here we have an important principle in mind. We should not let the water stand on the surface of the soil, completely ponded over the whole distance, but should let it go into the soil and then apply another application of non-saline water after the water midway between the drains has had a chance to fall. Maintaining the layer of ponded water on the surface prevents leaching midway between drains. This would seem to be contrary to what we would expect. Of course this is only true when there is an impermeable layer near the surface which stops water movement. If there were gravel or a coarse layer of shells below the soil, then the ponding action would be ideal as we saw in Fig. 15.

PROCESSES CAUSING PESTICIDE MOVEMENT IN SOIL

Miscible displacement, diffusion and leaching are physical processes involved in the movement of pesticides in soil. Dr. Cleve A. I. Goring of the Dow Chemical Company, Western Division, Seal Beach, California has supplied the following information. We are grateful to him for permission to quote it.

"In general, pesticides fall into three categories as far as movement in soil by water is concerned.

"1. Chemicals with a reasonably high water solubility, that are non-ionic (phenyl N, N'-dimethylphosphorodiamidate [Nellite^(R)], allyl alcohol, formaldehyde) or are organic acid salts where the toxicant portion is anionic. The latter would include the sodium or amine salts of dithiocarbamic and ethylenebis dithiocarbamic acids, 2,4-D, 2,4,5-T, chlorobenzoic trichloroacetic, 2,2-dichloropropionic, and other organic acids.

"2. Soluble organic salts where the toxicant portion is an organic cation, such as, quaternary ammonium germicides and pyridylum herbicides (Paraquat and Diquat).

"3. Fat soluble and/or highly water insoluble non-ionic chemicals such as 2-chloro-6-(trichloromethyl)pyridine (N-Serve^(R)), DDT, chlorinated hydrocarbon insecticides, urea and triazine herbicides, and fungicides such as pentachloronitrobenzene and captan.

"Chemicals with properties intermediate between groups also occur but can usually be assigned to one of the groups.

"Group (1) is not sorbed strongly by any of the soil constituents. I presume their movement through soil would follow the same rules as movement of, say, nitrate or chloride. I believe that various modifications of Day's (Day, 15) hydrodynamic dispersion theory would be applicable to their movement depending on the initial and final boundary conditions.

"Group (2) could presumably be treated somewhat in the same manner as Bower (14) treats movement of cations except that organic cations are usually much more strongly sorbed than inorganic cations.

"Group (3) is a more complex problem. For chemicals with relatively low water solubility and high fat solubility such as N-Serve one can calculate a partition coefficient for distribution between the water and the soil organic matter which contains fat (Goring, 19). The kind of mathematical treatment that might apply might be somewhat similar to that used by Bower for cations, by simply assuming an absorption equilibrium between water and the soil organic matter. One complication is that to reach this equilibrium all of the chemical applied to the soil must first dissolve in the soil water and soil organic matter. A long exposure period may be required for chemicals in this group to reach this type of equilibrium."

CONCLUDING COMMENTS AND SUMMARY

In conclusion, I would like to show you a curve (fig. 20) that has

Insert Fig. 20. Capillary diffusion and molecular self-diffusion coefficients plotted against each other for three soils. (Kunze and Kirkham, 27.)

really surprised us, which combines capillary diffusion and molecular

self-diffusion. This curve does not pertain to the leaching process, but to the movement by the two processes, capillary diffusion and molecular self-diffusion. Here we see that the capillary diffusion coefficient D_C and the molecular self-diffusion coefficient D_S , when plotted against each other for different soils, all lie on the same universal curve. Points on this curve are for a clay, a silt loam, and a silty clay loam, soils of widely different textures, and at widely different moisture contents; and yet when the curve of self-diffusion versus capillary diffusion is plotted, we get the universal (for three soils) curve shown. We have been able to show that these two diffusivities are theoretically related through the viscosity of the water; but beyond that we are not able to understand why we get the universal curve.

In summary: we have seen how water may move chemicals and pesticides in the soil by miscible displacement, diffusion, and leaching. We have noted a number of diffusion processes and have concluded by showing that the capillary diffusivity and the molecular diffusivity of soil water appear to be related by a universal curve for soils of wide textural differences. We have drawn attention to certain aspects of water movement which are not understood. To understand these aspects is a challenge for future exciting research.

ACKNOWLEDGMENT

I am indebted to John C. Corey, Research Assistant in Soil Physics, for help in preparing this paper.

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Figure 20. Capillary diffusion and molecular self-diffusion coefficients plotted against each other for three soils. (Kunze and Kirkham, 27.)

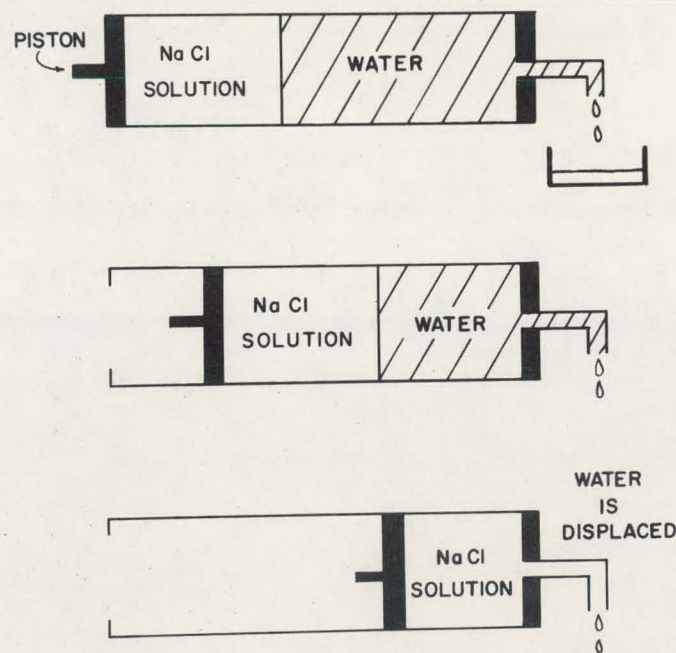


Figure 1. Piston flow in a tube.

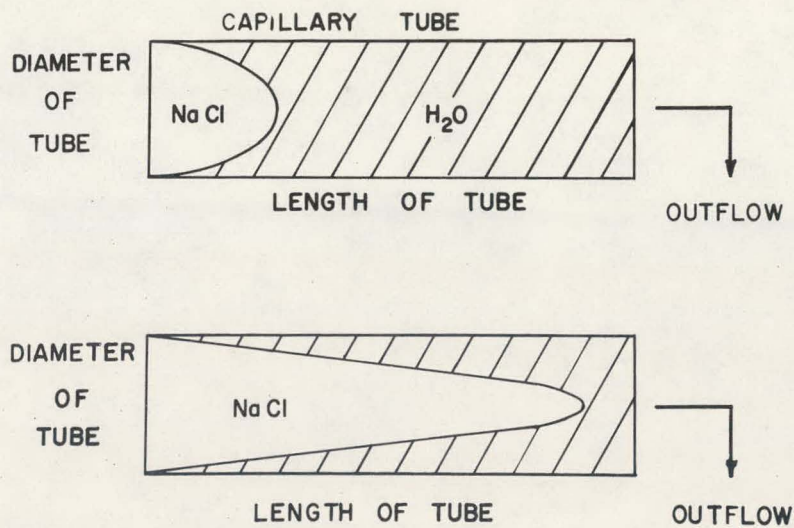


Figure 2. Capillary flow in a tube assuming no molecular diffusion. (Nielsen and Biggar, 20.)

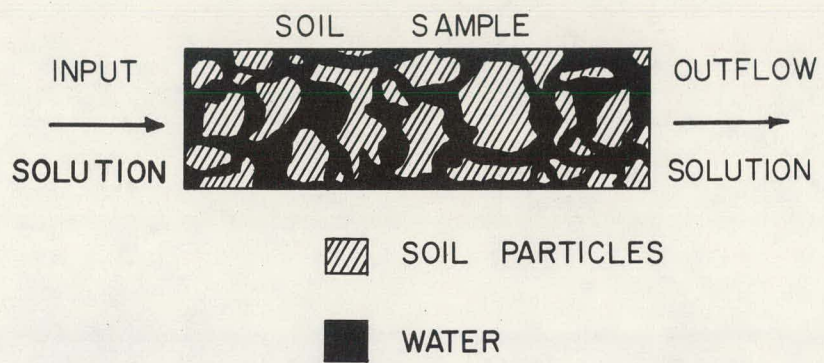


Figure 3. Flow in saturated soil.

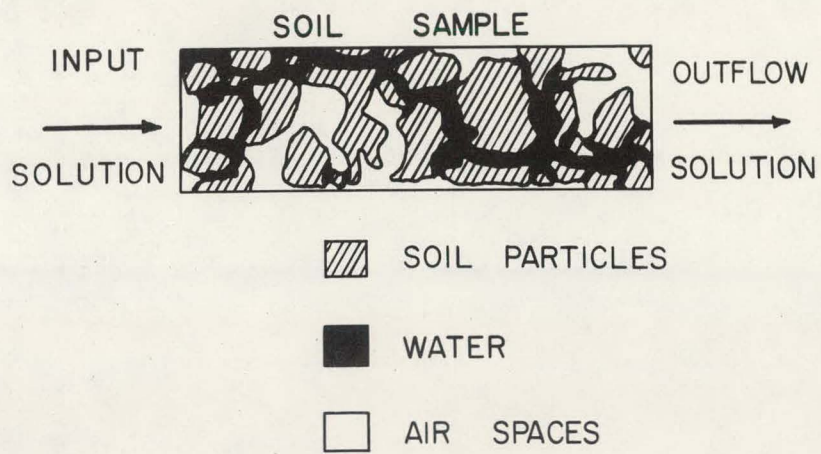
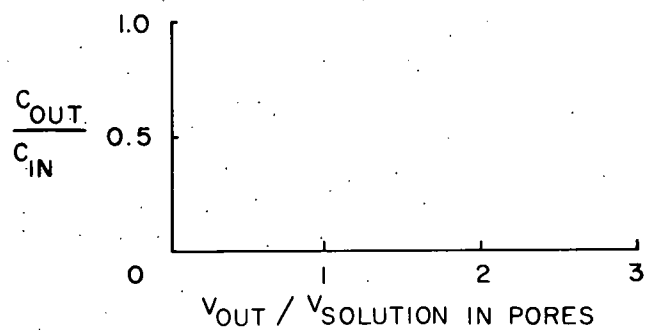


Figure 4. Flow in unsaturated soil.



C_{OUT} CONCENTRATION OF NaCl IN THE
OUTFLOW SOLUTION (gm/cc)

C_{IN} CONCENTRATION OF NaCl IN THE
INPUT SOLUTION (gm/cc)

V_{OUT} VOLUME OF OUTFLOW SOLUTION

$V_{SOLUTION\ IN\ PORES}$ VOLUME OF
WATER HELD
IN SOIL PORES OF THE SAMPLE
AT TENSION USED

Figure 5. Way of presenting miscible displacement data.

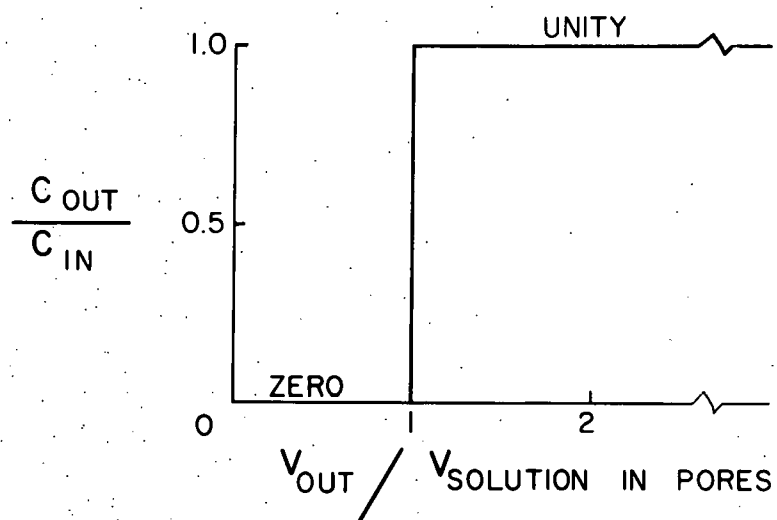


Figure 6. Outflow curve for piston flow. (Nielsen and Biggar, 20.)

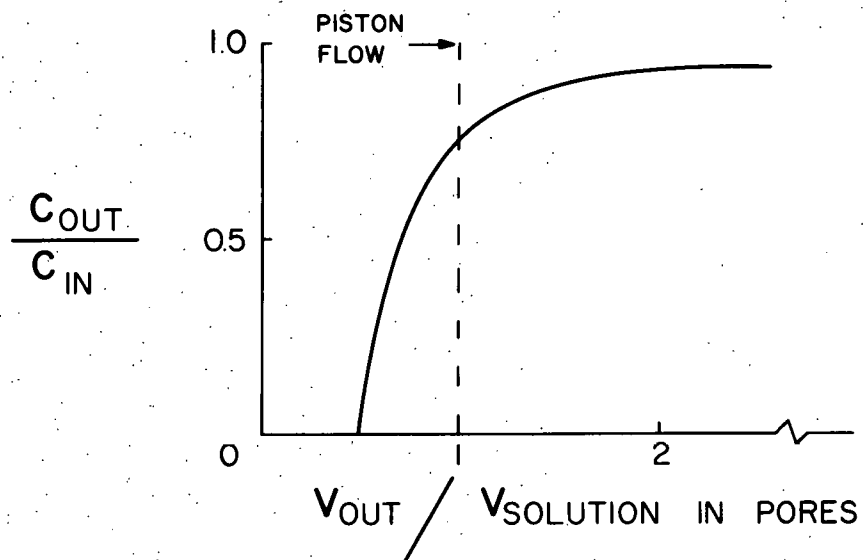


Figure 7. Outflow curve for capillary flow in a tube assuming no molecular diffusion. (Redrawn from Nielsen and Biggar, 20, their figure 7.)

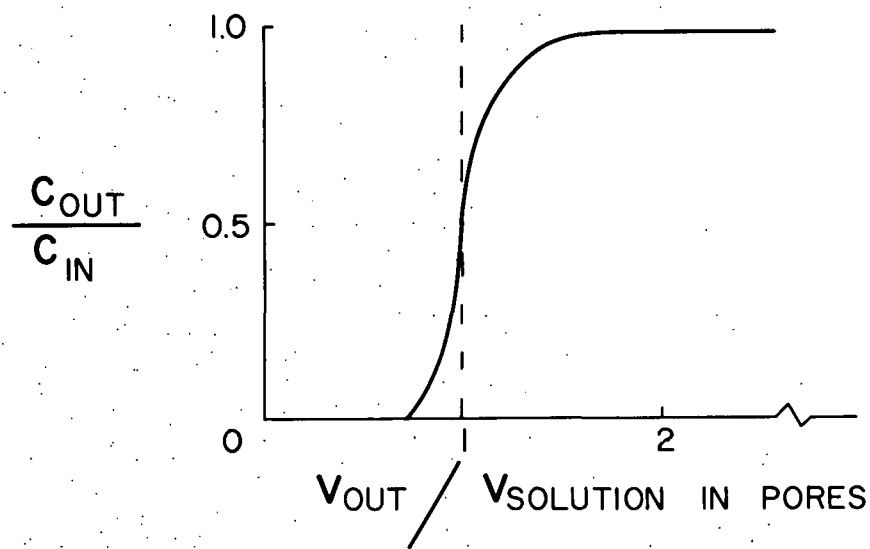


Figure 8. Outflow curve for flow in saturated soil. (Nielsen and Biggar, 20.)

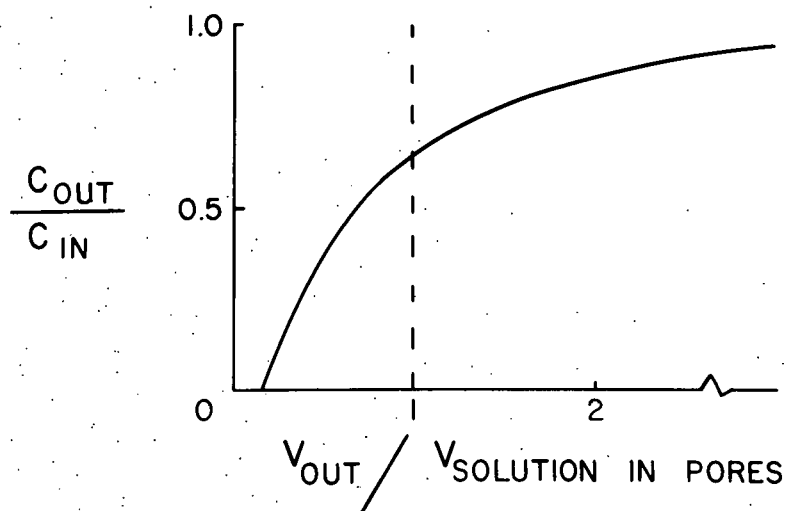


Figure 9. Outflow curve for unsaturated flow in soil.

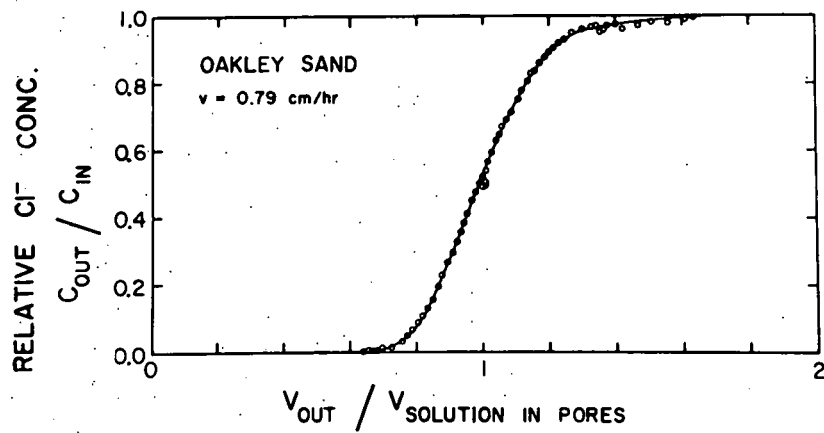


Figure 10. Outflow curve for chloride in Oakley sand. (Nielsen and Biggar, 20.)

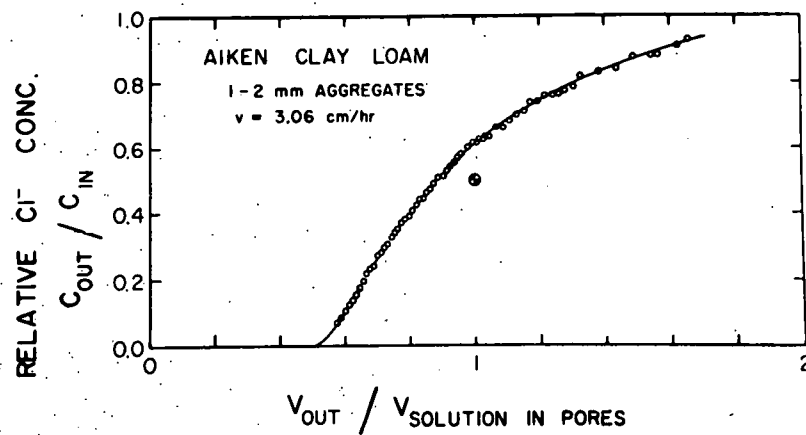


Figure 11. Outflow curve for chloride in Aiken clay loam. (Nielsen and Biggar, 20.)

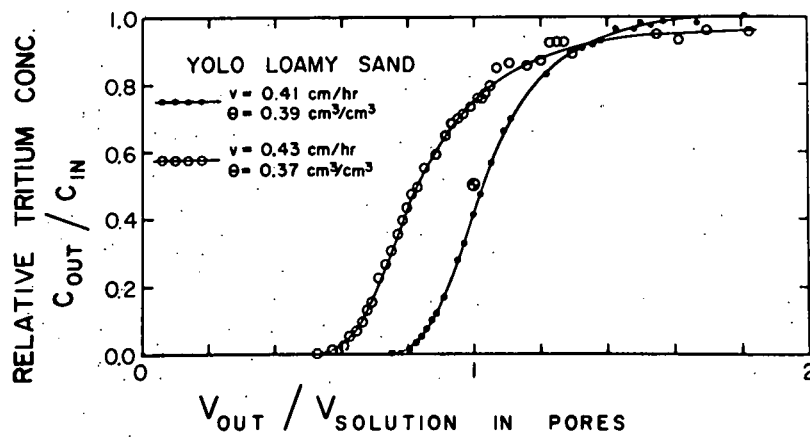


Figure 12. Outflow curve for tritium in Yolo loamy sand. (Nielsen and Biggar, 20.)

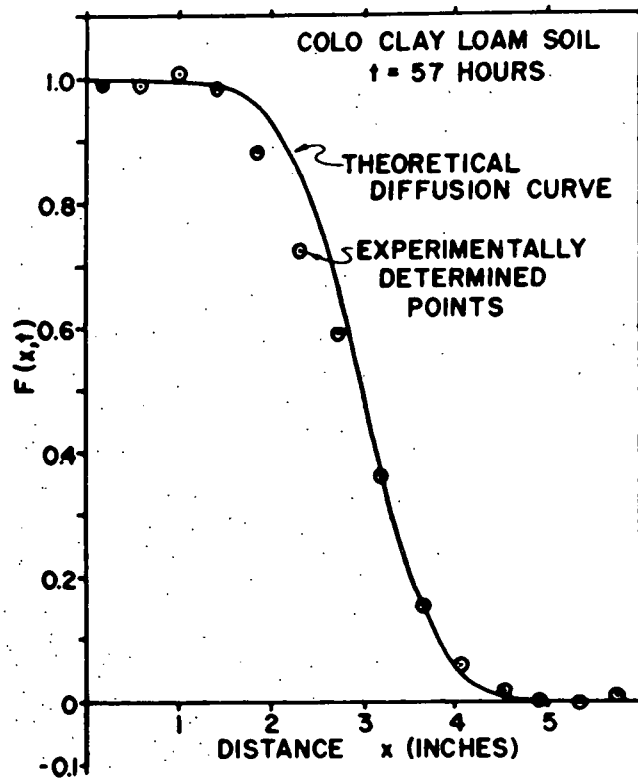


Figure 13. Molecular self-diffusion of water in Colo clay loam soil. (Kunze and Kirkham, 21.)

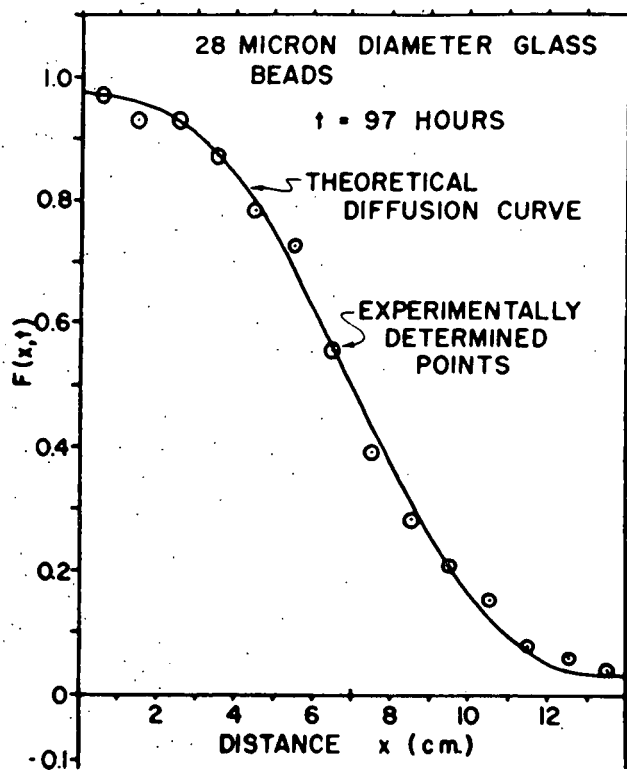


Figure 14. Molecular self-diffusion of water in glass beads. (Kunze and Kirkham, 21.)

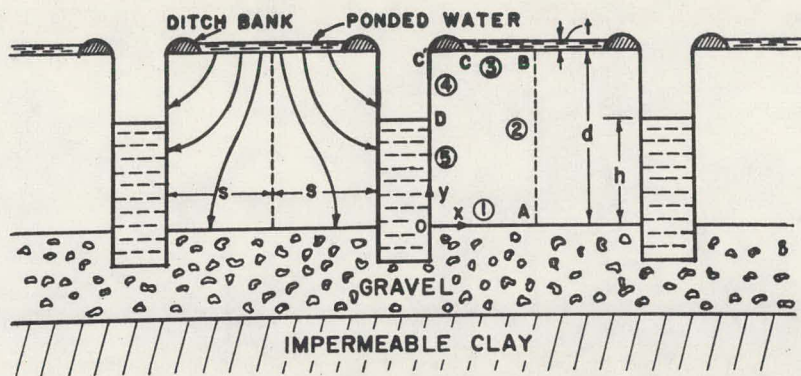


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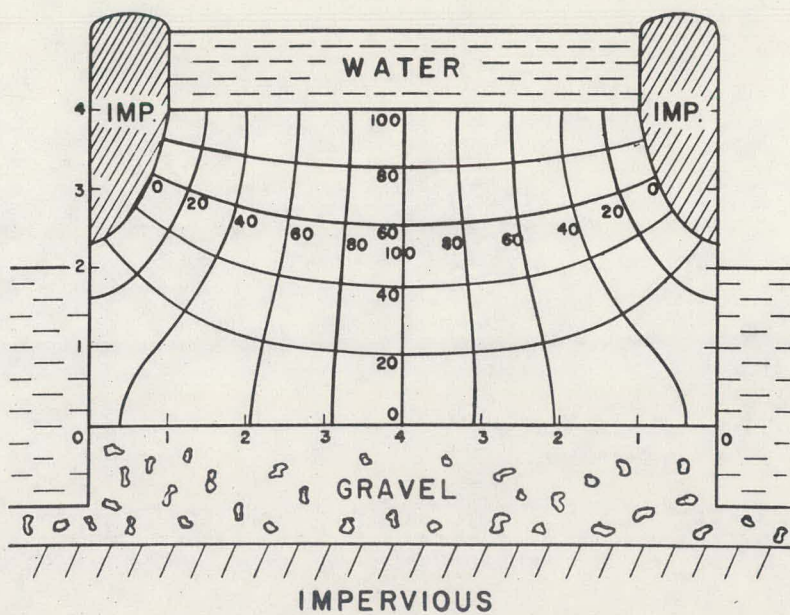


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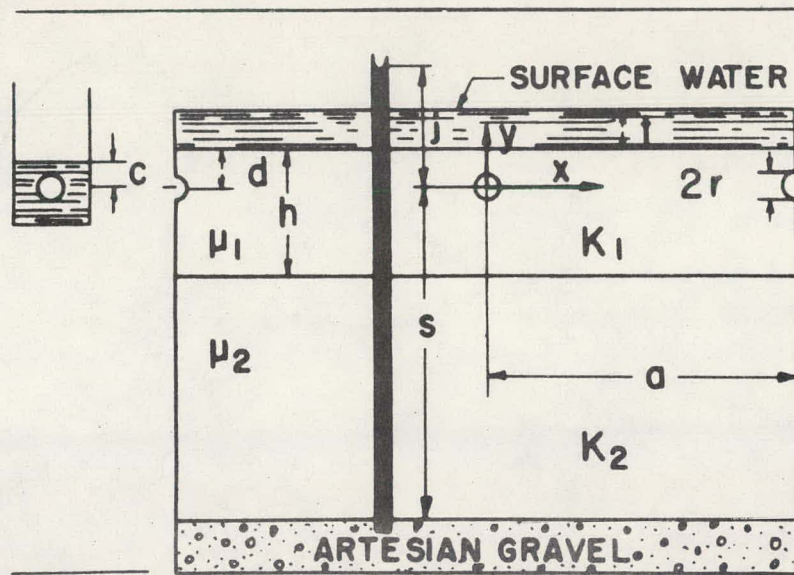


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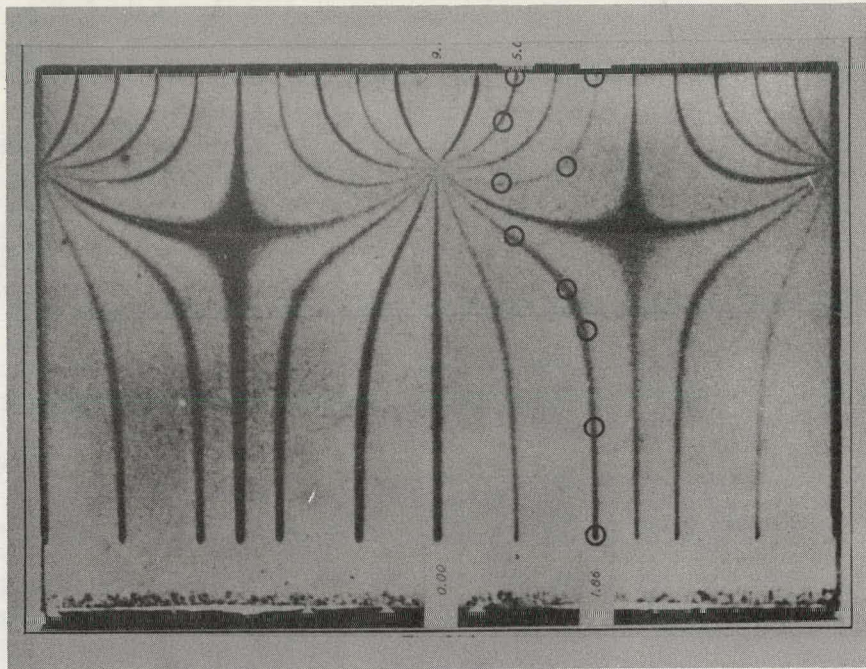


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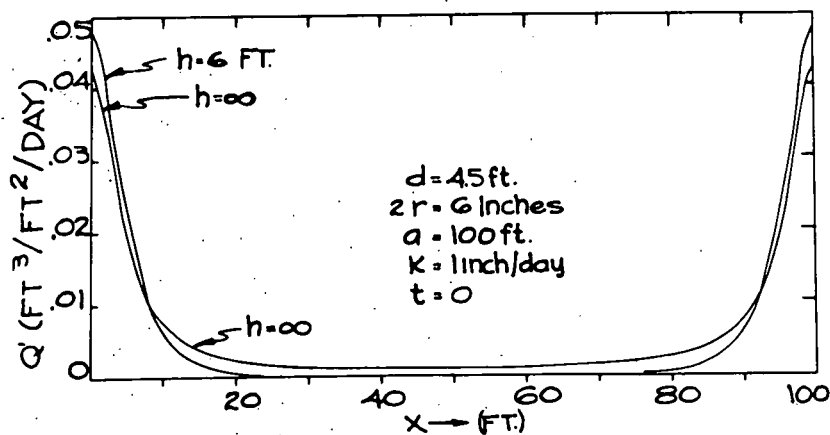


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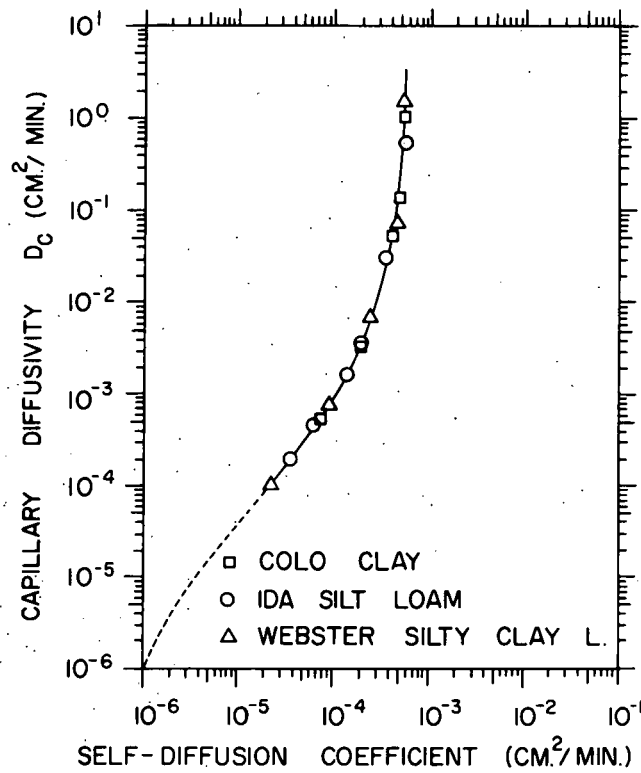


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