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EFFECT OF THE PRIOR DISPOSAL
OF UNCONTAMINATED SOLUTION
ON THE WASTE RETENTION CAPACITY
OF A HANFORD CRIB

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Disposal and Processing

EFFECT OF THE PRIOR DISPOSAL OF UNCONTAMINATED SOLUTION
ON THE WASTE RETENTION CAPACITY OF A HANFORD CRIB

By

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EFFECT OF THE PRIOR DISPOSAL OF UNCONTAMINATED SOLUTION
ON THE WASTE RETENTION CAPACITY OF A HANFORD CRIB

R. C. Routson

INTRODUCTION

This report presents the experimental results in an evaluation of the effect of the prior disposal of an uncontaminated solution upon the specific retention capacity of a Hanford crib. Specific retention is commonly defined as the maximum amount of water that a soil or sediment will retain against gravity drainage, and it is usually expressed on a volumetric basis. However, a somewhat modified concept of the use of the term specific retention capacity will be presented in this document.

Specific retention is the result of the interactions between the solid and liquid phases in porous media. Contact points between adjoining particles of porous media, such as soil, form v-like grooves which act as capillaries. The effective capillary size of these interstices varies from zero at the point of particle contact to much larger sizes as the distance from the contact increases. This results in a large range of effective capillary sizes. When initially dry, porous media will sorb water if placed in contact with liquid water or from an atmosphere containing water vapor. Water is sorbed due to the existence of adhesive forces between solids and water and cohesive forces between water molecules. When initially saturated, a porous media will drain as the system tends to come to equilibrium. At equilibrium all water is energetically equivalent and capillaries smaller than a determinable equivalent size will retain small quantities of water which collectively represents the specific retention capacity of a soil. Additional water is sorbed due to exchangeable ion and site hydration, but the contribution of such water to the specific retention capacity is probably small in low exchange capacity sands such as those found on the Hanford project. The maximum size of capillaries retaining water at equilibrium is a function of the physical and chemical nature of the porous media, the position of the porous media in a gravitational field with respect to a free water surface, temperature, pressure, and direction of approach to equilibrium. In cases where free water is not present the equilibrium is controlled by the relative

humidity of the atmosphere surrounding the porous media. Thus, theoretically, any porous media at equilibrium, following drainage, has some capacity for water retention against gravitational attraction to a free water table and vaporization. Practically, measurement of the specific retention capacity of soils and sediments is difficult because under most conditions water movement in soil is exceedingly slow and equilibrium can be achieved only after extended periods of time. These equilibrium times are extremely long with respect to times available for human measurement.

Two general types of techniques have been used to "measure" the specific retention capacity of porous media. These techniques are: 1) column, and 2) centrifuge moisture equivalent methods. In column techniques an initially saturated soil is allowed to drain to a simulated water table until equilibrium is achieved, and the moisture content or moisture distribution above the water table may be considered to be the specific retention capacity. In practice even low-capacity sands have been found to be draining at slow rates several years after the initiation of such experiments (King, 1899; Johnson et al., 1963). In order to avoid these long-term soil drainage experiments, centrifuge moisture equivalent techniques have been developed. In these techniques the soil moisture is acted upon by a centrifugal force many times the force of gravity under defined conditions to obtain a more rapid expulsion of water. Unfortunately, there is no known direct method of relating centrifuge moisture equivalent to the specific retention capacity of a soil. This has been discussed in some detail by Bierschenk (1959). However, the centrifuge moisture equivalent has been widely used as a quasiphysical index of the ability of porous media to retain water against gravitational forces, and some success has been reported in relating this measurement to the specific retention capacity of a soil if the definition of the specific retention capacity contains a real or implied time period for drainage (Piper, 1933).

Thus it would seem that reported specific retention capacities do not refer to equilibrium values, but to measured values at some nonequilibrium time when the rate of drainage becomes small. There is no proof that the ultimate (equilibrium) value will not be significantly smaller than the measured value.

It would seem that a specific retention drainage time should be chosen for each application which is commensurate with that application. For example, for an application involving a proposed pumping test, a specific retention time (used to determine specific yield) of a few days duration and accurate to a few percent would be adequate; while in contrast, for an application involving the ground disposal of long-lived radionuclides a specific retention drainage time of several half-lives of the radioactive wastes involved would seem more appropriate. In the above sense, specific retention capacity is a misnomer, and a term such as the water retention capacity of a soil after a specified time of drainage would be more appropriate.

With the above concepts in mind the effect of the previous disposal of an uncontaminated solution on the specific retention capacity of a soil was evaluated without measuring the specific retention capacity of the soils involved. In this study techniques were developed to evaluate the rate of movement of a pulsed waste front subsequent to addition of solution to a soil column, relative to the rate of movement of a similar pulsed waste front with no prior addition of solution. If, in this study, the rate of movement of the waste front was increased by the prior addition of solution, it was adduced as proof that the specific retention capacity (waste retention capacity) of that soil for waste solution was reduced; conversely, if the rate of frontal movement was decreased, the waste retention capacity of this soil was increased. In the remainder of this paper the term waste retention will be used in lieu of specific retention capacity. Waste retention capacity is defined as the amount of aqueous waste solution which a soil column will retain after a specified time such that the waste front just intercepts a real or assumed water table.

Summary

An empirical method, suggested by constant head and moisture redistribution studies, was developed to predict the movement of a wetting front and a waste front in homogeneous sand columns due to pulse solution additions. The movement of wetting fronts and waste fronts were initiated by additions of equal pulses of aqueous solution. Movement of the wetting front was initiated prior to the initiation of the movement of the waste front.

Log-log plots of the frontal position versus elapsed time were found to be linear for both wetting fronts and waste fronts for elapsed times up to 5000 hours. This demonstrates that the position of a front at time T can be empirically predicted by equations of the form $S = ET^F$. Parameters E and F can be evaluated from laboratory measurements. Parameter E was found to be an extensive variable which increased with an increase in the amount of solution added as a pulse. Parameter F was found to be constant for a range of solution additions from 2.5% to 10% of the column volumes, suggesting that it may be an intensive variable.

The position of the wetting front was found to move at a faster rate than the position of the waste front. Initiation of the waste front was found to increase the rate of movement of the wetting front. The observed decreased rate of movement of the waste front relative to the wetting front was interpreted as being equivalent to an increase in the waste retention capacity of the soil due to an initial addition of uncontaminated solution to the soil. In addition, moisture initially present in a soil need not be subtracted from the waste retention capacity because it will be displaced into the ground water ahead of any disposed aqueous waste.

Materials and Methods

Six vertical 3.7 cm diameter by 100 cm long glass columns were hand-packed with an air-dry Burbank s. subsoil to a bulk density of 1.6 g/cm^3 . Care was taken to pack the columns to homogeneous bulk densities. The bottom of each column was open to the atmosphere through a pin hole. Each column was taped with black electrical tape to exclude light and limit the growth of microorganisms. A saturated atmosphere was maintained above each column following solution addition.

A 3400 ppm disodium EDTA solution was prepared and adjusted to pH 1.0 with HCl. This solution will, henceforth, be referred to as the EDTA solution. Preliminary experiments were conducted to test the feasibility of tracing the movement of a waste front with an EDTA- ^{60}Co complex.

In the first preliminary experiment, the stability of the EDTA- ^{60}Co complex was tested as a function of pH at values of 1.0, 5.0, and 8.2. The EDTA solution was spiked with ^{60}Co and was adjusted to pH 5.0 and 8.2 with

NaOH. A liter of each of the EDTA-⁶⁰Co solutions was continuously pumped through small soil columns containing 2.0 gms of a Burbank s. subsoil. Sorption of ⁶⁰Co on the soil was continuously monitored by counting the 1.33 mev γ -peak, using a 4.10 AS Packard, single-channel, Auto-Gamma Spectrometer with a NaI scintillation crystal detector.

In the second preliminary experiment, the top 12.0 cm of a 100 cm column was packed with a Burbank s. subsoil to which 5% by volume of the EDTA solution spiked with ⁶⁰Co was added. The remaining 88 cm of the column were air filled. Thorough mixing of the solution and soil was accomplished prior to packing the column. In the column the abrupt end of the moistened-spiked soil should simulate a sharp wetting front. The distribution of detected activity as a function of distance was made by directly counting the column with the spectrometer previously described. A tunnel with a 0.4 cm counting slit was constructed of lead bricks for accommodating the column during the measurement. Detected activity was the number of disintegrations detected by the spectrometer per unit time without correction for geometry, efficiency, or sorption. Distance was measured from the top of the soil with meter sticks fastened to the sides of the Pb-tunnel used to shield the columns.

Six columns were given three different solution treatments in duplicate. To columns 1 and 2, 10% by column volume of the EDTA solution spiked with ⁶⁰Co was added as a pulse, and the movement of the wetting front as a function of time was measured by both visual and tracer methods previously described. To columns 3 and 4, 5% by column volume of the EDTA solution was added as a pulse and the movement of the wetting front was measured visually. At a subsequent time a second 5% by column volume of the EDTA solution spiked with ⁶⁰Co was added as a pulse and the movement of the waste front was measured as a function of time by the tracer technique. Time was measured from the time of solution additions and were designated T_{01} and T_{02} for the initiation of the first and second fronts, respectively. Columns 5 and 6 were treated exactly like columns 3 and 4 except 2.5% by column volume of the EDTA and spiked EDTA solution were added, respectively.

Results and Discussion

Properties of the Burbank s. subsoil used in the experiment are given in Table 1. This soil is a slightly calcareous, low-exchange capacity sand which is somewhat typical of the glaciofluvial sediments found on the Hanford project to which radioactive waste solutions are disposed.

In the first preliminary experiment no sorption was detectable at pH 1.0, 5.0, and 8.2. In contrast, at pH 5.0 greater than 10,000 cpm would be detected for a small column at equilibrium if the ^{60}Co were not complexed. It was anticipated that the solution pH would be between the initial pH of 1.0 and 8.2. Thus, the EDTA- ^{60}Co complex was deemed to be stable at solution pH expected in the soil column.

Due to the energy of the ^{60}Co - γ , the thickness of lead necessary to obtain perfect shielding would be prohibitively large. Thus, it was necessary to ascertain if the position of the wetting front could be located with sufficient precision by measuring the detected activity, using the described experimental apparatus. In the preliminary column containing 12.0 cm of moistened-spiked soil, the distribution of detected activity as a function of distance is given in Fig. 1. It can be seen that the end of the spiked soil can be detected and is at the inflection point of the detected activity-distance curve. This suggests that the position of a front may be detectable by determining the inflection point of a detected activity-distance curve.

In columns 1 and 2, in which the wetting and waste fronts should be coincident, the average distribution of detected activity as a function of distance is given in Fig. 2 after 772 hrs. It is evident that after 772 hrs the position of the wetting and waste fronts is coincident and can be determined either visually or by determining the position of inflection point of the detected activity-distance curve. Table 2 is a listing of the position of the wetting front as determined by both visual and inflection point methods. In no instance do the two measurements differ by more than 0.5 cm which is considered to be the accuracy of the method. This experiment demonstrates that the frontal position can be determined by the tracer-inflection point technique.

While little direct research has been conducted on the frontal movement of pulses of aqueous solution, constant head and moisture redistribution

Table 1

Properties of Burbank s. C horizon

% sand	Particle size		pH in 0.01 CaCl ₂ units	CaCO ₃ mg/g	CEC meq/100 g
	% silt	% clay			
89.4	10.1	0.5	7.1	0.79	1.9

FIGURE 1. Detected Activity Along a 100 cm Column with the Initial 12.0 cm packed with a Burbank s. Subsoil Which had been Thoroughly Mixed with 5% by Volume of EDTA ^{60}Co Solution. The end of the Soil Simulates a Wetting Front (12.0 cm).

FIGURE 2
Average Detected Activity from Columns 1 and 2 at 772 Hours.
The Visual Wetting Front and Inflection Point were Coincident
at 88.4 cm

Table 2

Average Movement of the Wetling Front of Columns 1 and 2

Average Movement of the Wetling Front of Columns 1 and 2			
Sampling	Position of Wetling Front (Visual)		Elapsed Time (hrs)
	Position of Wetling Front (Inflection point)		
	(cm)		
1	61.4	61.5	24
2	66.0	65.5	48
3	69.5	69.0	96
4	74.7	74.5	168
5	75.1	75.0	192
6	75.8	76.0	216
7	76.3	76.0	240
8	79.2	79.5	336
9	80.6	80.5	384
10	83.0	83.0	504
11	88.4	88.5	772
12	90.9	90.5	988

studies do suggest the possibility that an empirical equation of the form

$$S = ET^F \quad [1]$$

may describe the movement of pulsed fronts as a function of time; where, S is the distance of the wetting front from the soil surface at elapsed time T ; and E and F are empirical parameters which must be evaluated for each soil. If equation [1] is satisfied, a log-log plot of frontal position versus elapsed time will be a straight line. Parameter F is equal to the slope of this straight line, and parameter E is equal to the frontal position at T equal to 1.

Log-log plots of the average frontal position versus elapsed time for the Burbank s. subsoil are given in Fig. 3 for columns 1 and 2, 3 and 4, 5 and 6. It can be seen that the movement of all fronts do give linear log-log plots demonstrating the validity of the use of equation [1] over the measured time period. It can also be seen that the movement of the waste fronts, which were initiated subsequent to the wetting fronts, is at a slower rate than the wetting fronts. This is demonstrated by the significantly decreased slope of the waste fronts relative to the slope of the wetting fronts. Fig. 4 is a linear plot of wetting and waste frontal positions versus elapsed time for columns 5 and 6. It can be seen that the slope (instantaneous rate of movement) of the waste front is less than the slope of the wetting front at all plotted times.

Table 3 gives the square of the least squares regression coefficients, standard errors of the estimate, and calculated values of parameters E and F for the frontal data plotted in Fig. 3. The slope of the wetting fronts (F) of all three sets of columns were found to be equal within experimental error. This was true even though the volume of solution added to initiate the wetting frontal movement in columns 1 and 2 was four times the volume added to columns 5 and 6. The values of F for the waste fronts may also be equal, but insufficient data are available to completely evaluate this possibility.

Addition of solution to initiate the movement of the waste fronts greatly increased the rate of movement of the wetting fronts. This is demonstrated by a greater than a three-fold increase in slope of the curve from 0.1 to 0.34 for the first fronts of columns 5 and 6 subsequent to the waste front solution addition.

FIGURE 3
Average Movement of Pulses of Aqueous Solution in 100 cm Burbank s Columns

FIGURE 4
Average Position of a Wetting and Waste Front Added to Columns 5 and 6 at
Different Times as Function of Time. The Waste Front (Second Front) was Added
at Time T_{02} After the Wetting Front

Table 3
Least Squares Regression Analysis of Frontal Movements in 100 cm Burbank s. Columns

Column No.	treatment	front number	r^2	Standard Error of Estimate $(S^2)^{1/2}$	E	F
1 and 2	10% solution	1	0.972	0.008	41.5	0.10
3 and 4	5% solution	1	0.982	0.006	17.3	0.10
	+5% solution	2	0.935	0.008	19.5	0.07
5 and 6	2.5% solution	1	0.976	0.009	9.2	0.11
	+ 2.5% solution	1*	0.989	0.013	2.0	0.34
		2	0.906	0.010	9.2	0.08

*Data are for the wetting front subsequent to addition of solution to initiate the waste front.

The value of parameter E was found to increase as the amount of solution added as a pulse increased. An increase of 9.2 to 41.5 was observed in this experiment upon initial solution additions of from 2.5% to 10%, respectively. Thus it would seem that the value of parameter F may be a function of a soils texture, structure, and composition (intensive variable for a given solution). The value of the parameter F (0.10) is only about one fifth of the value found by Kirkman and Feng (1949) for a frontal movement in their constant head studies. The consistently high values of the square of the least squares regression coefficients suggest that the technique discussed here is experimentally valid.

The observed decrease in the rate of movement of the waste front relative to the rate of movement of the wetting front is adduced as proof of an increase in the waste retention capacity of the soil as was discussed in the introduction. Furthermore, moisture initially present in the soil need not be subtracted from the waste retention capacity of the soil since this moisture will be displaced into the ground water ahead of any waste front and not increase the rate of waste movement.

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