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SUBSURFACE MOVEMENT OF RADIONUCLIDES

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by

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SUBSURFACE MOVEMENT OF RADIONUCLIDES

In this presentation we shall be concerned with how radionuclides move in the subsurface. This is the realm of groundwater flow; so we shall spend some time examining the subsurface movement of water before the migration of radionuclides is considered. We will consider the source of groundwater, what forces make it move underground, the physical properties of aquifers, how to determine the direction and rate of flow, and finally the effects of dispersion and adsorption.

The source of most groundwater is the hydrologic cycle (Figure 1). Evaporation from the oceans and other secondary sources puts water vapor into the atmosphere which later falls as precipitation. A portion of this precipitation infiltrates below the land surface and becomes part of the subsurface water flow regime. The amount that infiltrates depends on many factors. A few of the most important are: the amount of rainfall, the rate of rainfall, soil type, and previous soil moisture.

The forces that make water move underground are mainly two: pressure and gravity or elevation. Figure 2 shows schematically how pressure and elevation can be used to move water. The sum of these two quantities is given the name hydraulic head (Figure 3). To measure hydraulic head in the field we simply drill a well and measure the water level in the well with regard to some datum (usually sea level, see Figure 3).

In 1856, in Dijon, France, an engineer named Darcy observed that water moves from areas of high head to areas of low head. He also observed that the amount of water moved was proportional to the head difference. These observations are today called Darcy's Law (Figure 4). In addition to the head difference between two points, the velocity of water flow depends on the hydraulic conductivity and the porosity of the aquifer. These are physical properties which will be discussed later.

There are two major classifications of aquifers: Confined and unconfined (Figure 5). An unconfined aquifer has a water table at atmospheric pressure. If a well is drilled into the unconfined aquifer the water in the well stands at the level of the water table. On the other hand, a confined (or Artesian) aquifer is bounded on the top and bottom by a relatively impermeable layer such as clay or shale. If a well is drilled into this aquifer the water level or hydraulic head will be above the upper confining layer. Confined and unconfined aquifers may both be present at a given location; the water level in the confined aquifer well may be above or below the water table, depending on the direction of water movement.

The unsaturated zone exists from the ground surface down to the water table (Figure 6). It is this zone that infiltrating water must traverse to become part of the water table and the saturated zone. The amount of moisture in this zone depends on the precipitation history as well as the physical properties of the zone.

We next turn our attention to the physical properties of the fluid and aquifer that effect groundwater flow. The porosity (Figure 7) of an aquifer is defined as the amount of void space in a sample divided by the sample volume. Typical porosities vary from about 1% to 50%. Porosities can be expressed as either percents or decimal values. Obviously, the void space of the porosity is the volume occupied by groundwater.

The hydraulic conductivity (Figure 8) depends on both fluid and aquifer properties. The main aquifer dependence is on the square of the average grain diameter. This means that fine grained materials will have low hydraulic conductivity and will not allow water to move through easily. There is an additional aquifer dependence, C, which depends on grain shape and pore interconnectivity; however, it is a complex function and can not be easily calculated for real aquifer materials. The fluid properties of weight density, γ, and viscosity, μ, also enter the expression for hydraulic conductivity. Fluids with higher weight densities and lower viscosities have greater hydraulic conductivity.

The specific yield (Figure 9) of an unconfined aquifer is defined as the amount of water that can be drained by the action of gravity from a unit volume. Obviously the specific yield must be less than the porosity because some water is held very tightly by molecular forces.

The storage coefficient (Figure 10) is defined for confined aquifers. It is the amount of water yielded by elastic properties per unit volume of the aquifer when the hydraulic head is lowered. It depends on the compressibility of water and the aquifer matrix, so it is much smaller than specific yield. For this reason, unconfined aquifers are generally much more prolific.

Figure 11 shows schematically some typical aquifer materials and a range of physical parameters for some typical geologic materials. Sketch (a) shows well sorted sand or gravel with good porosity and permeability. Sketch (b) shows a poorly sorted unconsolidated material with much lower porosity and permeability than (a). Sketch (c) is a well sorted unconsolidated granular material whose grains are also porous. Thus one would expect (c) to have even higher porosity than (a). Sketch (d) represents something like a sandstone which has its grains cemented together. One would expect (d) to have much lower porosity and permeability than (a). Sketch (e) represents a limestone. Usually the original porosity of limestone is low; however, fracturing and dissolution can cause secondary porosity. Sometimes this can be very pronounced as in areas where caves are very common. Sketch (f) represents something like crystaline rock that had little initial porosity but has been fracturecd to create secondary porosity and permeability. We do not have a sketch representing clay; but, it is very fine grained with a high porosity and very low specific yield and permeability. Shale also has a low permeability. Clays and shales are some of the main confining layers for confined aquifers.

One of the main tools of the groundwater hydrologist is the hydraulic head map. If one wishes to monitor the groundwater moveout in an area, the usual procedure is to drill a number of observation wells and record the water level in each. Each of these measurements can be located on an areal map. Lines of constant hydraulic head can be drawn through this data either by hand or computer program. Such a map is shown in Figure 12. Darcy's law tells us that water flows only in response to a change in hydraulic head. Therefore, water will flow perpendicular to the lines of constant head. Two sample flow lines are drawn on Figure 12. With this technique we can determine the direction a pollutant will migrate. With this knowledge it is possible some remedial action can be taken.

Since the Chernobyl accident occured in a river valley setting, I have decided to illustrate some of the things we have discussed by looking at the Kansas River valley near Lawrence (Figure 13). F - F' is a typical cross section of the river valley just north of Lawrence. The saturated thickness averages about 40 feet. It is an unconfined aquifer composed of unconsolidated sand and gravel with some clay and silt that has been deposited by river action (alluvium) over the recent geologic past (about one million years). We would expect the alluvium to have relatively high porosity and permeability. Beneath the alluvium, the bedrock consists of consolidated rocks that are relatively impermeable. This area gets about thirty five inches of rain on average each year. Some of this makes its way down to the water table and becomes part of the groundwater flow. The hydraulic head contours indicate that the water flow is mainly down the axis of the river valley. The river meanders back and forth across the valley. In some places the river gains water from the groundwater system; in other areas it loses water to the groundwater system. Overall the stream gains water from the groundwater system. This overall gain of water represents the amount of precipitation making its way to the river through the groundwater system. This type of behavior is usually called stream-aquifer interaction. Obviously, after heavy precipitation when the river stage is very high due to overland runoff, the river is feeding water into the groundwater system.

We have already considered Darcy's Law which will allow us to calculate the average groundwater velocity. The basic equation is shown again at the top of Figure 14. Using some typical values for hydraulic conductivity, porosity and hydraulic gradient for Kansas we can calculate some typical water velocities in different geologic environments. The major unconfined aquifers in Kansas are the Ogallala and the major river valleys. Typical water velocities in these unconfined aquifers would be 1-2 ft./day. In the central Kansas Lower Cretaceous aquifer consisting of the Cheyenne and Dakota Sandstones, typical water velocities would be .1-.01 ft./day. In clays and shales which have very low permeability, typical water velocities would be .01-.0001 ft./day. We see that even in prolific aquifers the water velocity is not large. For example in the Kansas River valley, on average it would take a water molecule 7-15 years to travel one mile.

In the preceeding discussion I have been careful to say average velocities. In actual fact, a water molecule may experience a range of velocities in moving from point A to point B. This is because the maximum velocity occurs at the center of a pore channel and goes to zero at the pore boundary (Figure 15). Also there are numerous ways a particle can go from A to B depending on which pore channel it traverses. If the water contains a solute whose concentration varies over space, molecular diffusion will cause the solute to be spread from areas of greater concentration to areas of lower concentration. Molecular diffusion will be operative until there is a uniform concentration throughout the aquifer. The cumulative effect of all these mechanisms is called hydrodynamic dispersion. The net effect of hydrodynamic dispersion is that a group of particles which started out together at point A will arrrive a point B at differing times.

Figure 16 shows a schematic for an experiment that would exhibit hydrodynamic disperison. If at some time t_0 we start feeding water with a tracer concentration C_0 through a sand colunm, it will be observed that the outflowing water will slowly build up to concentration C_0. If no dispersion was occuring the water would suddenly change from zero concentration to C_0.

Many solutes that are dissolved in groudwater react with the porous medium and are adsorbed or precipitated. Experimentally it is found that many times the mass of solute adsorbed or precipitated per unit mass of porous medium, S, is directly related to some power, b, of the concentration C (Figure 17). If b = 1, the solute is said to exhibit a linear isotherm. For the case of solutes which exhibit fast reversible reactions and linear isotherms, a retardation equation can be written involving the distribution coefficient, Kd (Figure 17). This means that the solute does not move as fast as the average water velocity and is retarded. Plugging typical values in for density and porosity, we find that when the distribution coefficient Kd = 1, the average solute velocity is only 1/5 to 1/10 that of the average water velocity. As Kd gets larger the solute becomes more immobile. If the distribution coefficient Kd = 100, the solute velocity is 1/400 - 1/1000 of the average water velocity

Figure 18 shows some experimentally determined distribution coefficients for common radionuclides in alluvium and bentonite clay. Fortunately, most of the Kd are quite large and consequently the movement of the nuclide will be severly retarded. ^{137}Cs and ^{90}Sr are usually considered the most environmentally troublesome. We see that ^{90}Sr is the more mobile in groundwater of the two. Figure 19 shows some data on the movement of radionuclides from a disposal crib for low level wastes at Hanford, Washington. In this case ^{106}RU was the most mobile, ^{90}Sr was the next most mobile, and ^{137}Cs was relatively immobile. This agrees well with the values for Kd that we saw in Figure 18.

Probably the best way to keep contamination of aquifers to a minimum in the event of a nuclear reactor accident is to have a containment vessel as an integral part of the design. However, if it is breached or absent, there are some other things that should be considered when the reactor site is selected. If possible, the reactor should not be sited over or near prolific aquifers, especially if they have a strong interaction with a nearby major stream. However, since the reactor needs a reliable supply of water for cooling and steam generation, this criterion for prevention of water pollution by an accident may be ignored.

If it is necessary to site a reactor over an aquifer, several other factors may be considered to keep water pollution to a minimum in case of an accident. A site should be selected that has a maximum depth to the water table. Clay-bearing geologic materials are effective in retarding radionuclide movement. A site should be chosen such that the geologic materials under the reactor have a large Kd. For example, in a river valley setting, a site with very clean sands and gravels and little clay would not be the best site from the standpoint of preventing radionuclide migration. Since a difference in hydraulic head is what causes water to move, a site should be chosen that has nearly a constant hydraulic head across it. If the geologic materials also have a low hydraulic conductivity, these two criteria will insure very low water velocities and, consequently, even lower rates of migration for radionuclides.

In summary, water velocities are usually low and radionuclide migration is usually retarded considerably. However, in cases where large quantities of radionuclides with relatively long half-lives such as ^{90}Sr are released, it is possible to have significant contamination of the groundwater system and any strongly coupled nearby surface water body. We have discussed several factors that, if considered at the time the reactor site is selected, could minimize the amount of water contamination resulting from an accident.

BIBLIOGRAPHY

Davis, S.N., and DeWiest, R.J.M., 1966, Hydrogeology, John Wiley and Sons, Inc., New York, p. 129-155.

Freeze, R.A., and Cherry, J.A., 1979, Groundwater, Prentice-Hall, Inc., Englewood Cliffs, N.J., p. 402-454.

Geophysics Study Committee, 1984, Groundwater Contamination, National Academy Press, Washington, D.C., p. 104-108, p. 139-146.

Todd, D.K., and McNulty, D.E.O., 1976, Polluted Groundwater, Water Information Center, Inc., Post Washington, New York, p. 41-47.

Siegel, F.R., 1974, Applied Geochemistry, John Wiley and Sons, New York, p. 293-316.

Walton, W.C., 1970, Groundwater Resource Evaluation, McGraw-Hill Book Co., New York, p. 29-206.

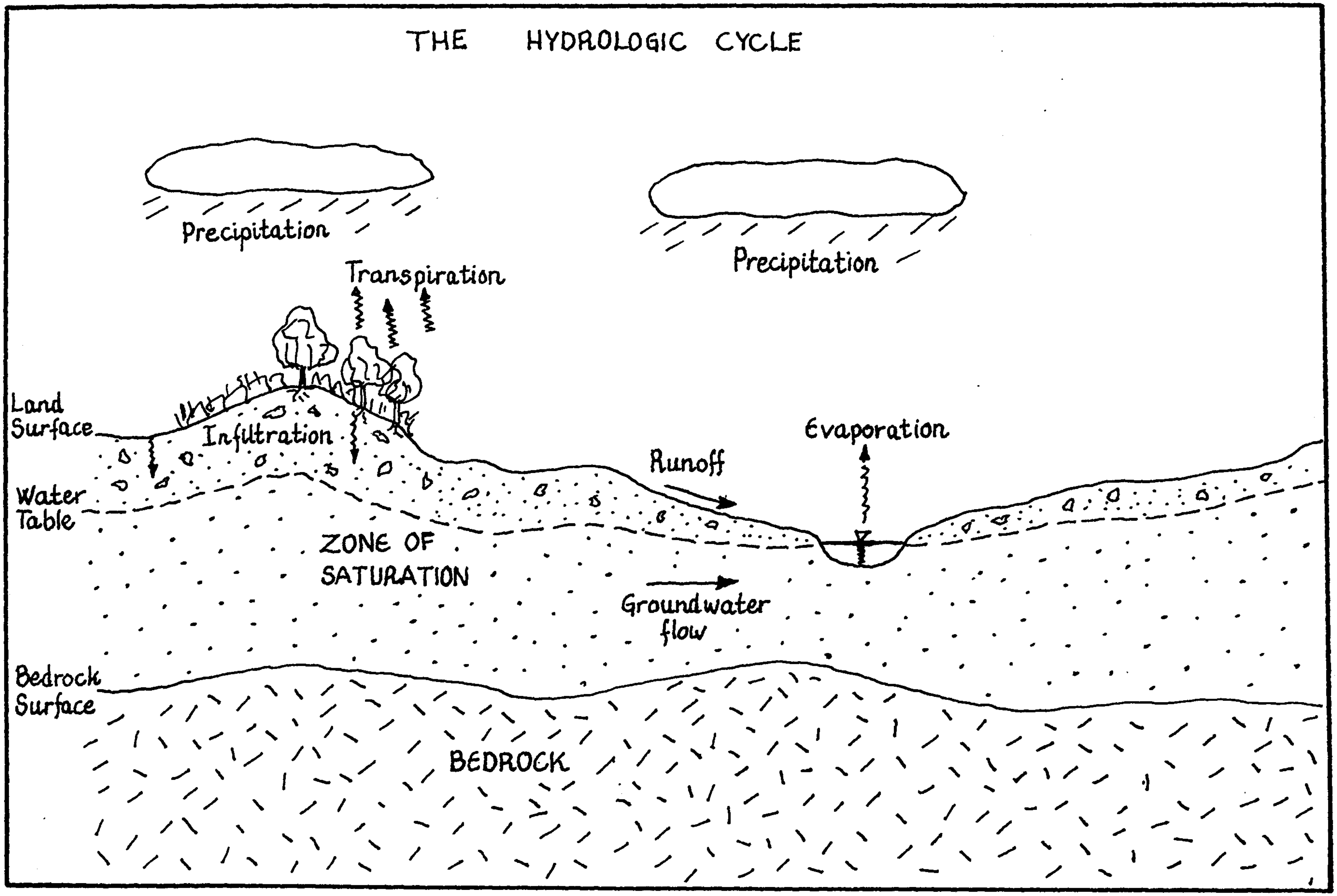

THE HYDROLOGIC CYCLE
Precipitation
Precipitation
Transpiration
Land Surface
Infiltration
Evaporation
Runoff
Water Table
ZONE OF SATURATION
Groundwater flow
Bedrock Surface
BEDROCK

Figure 1

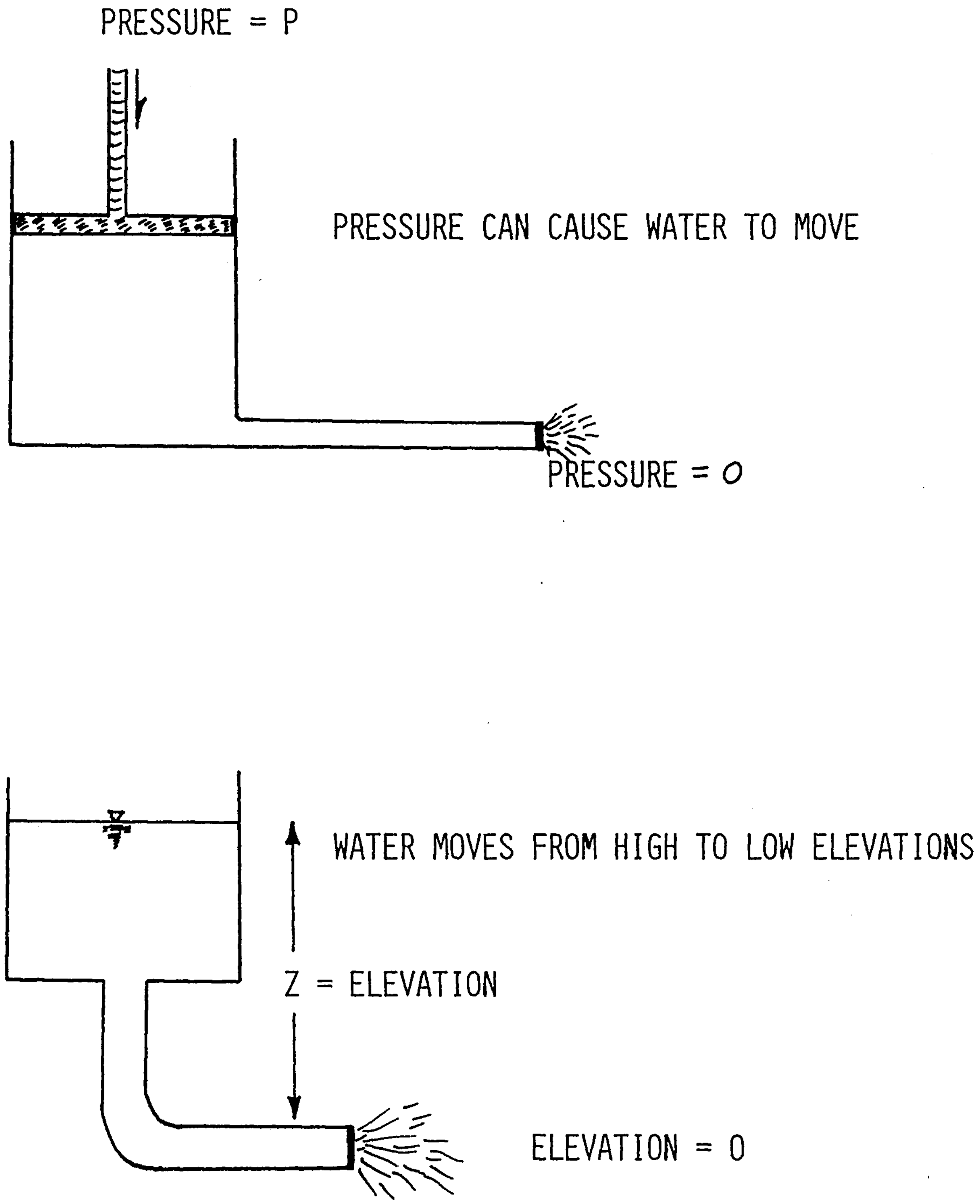
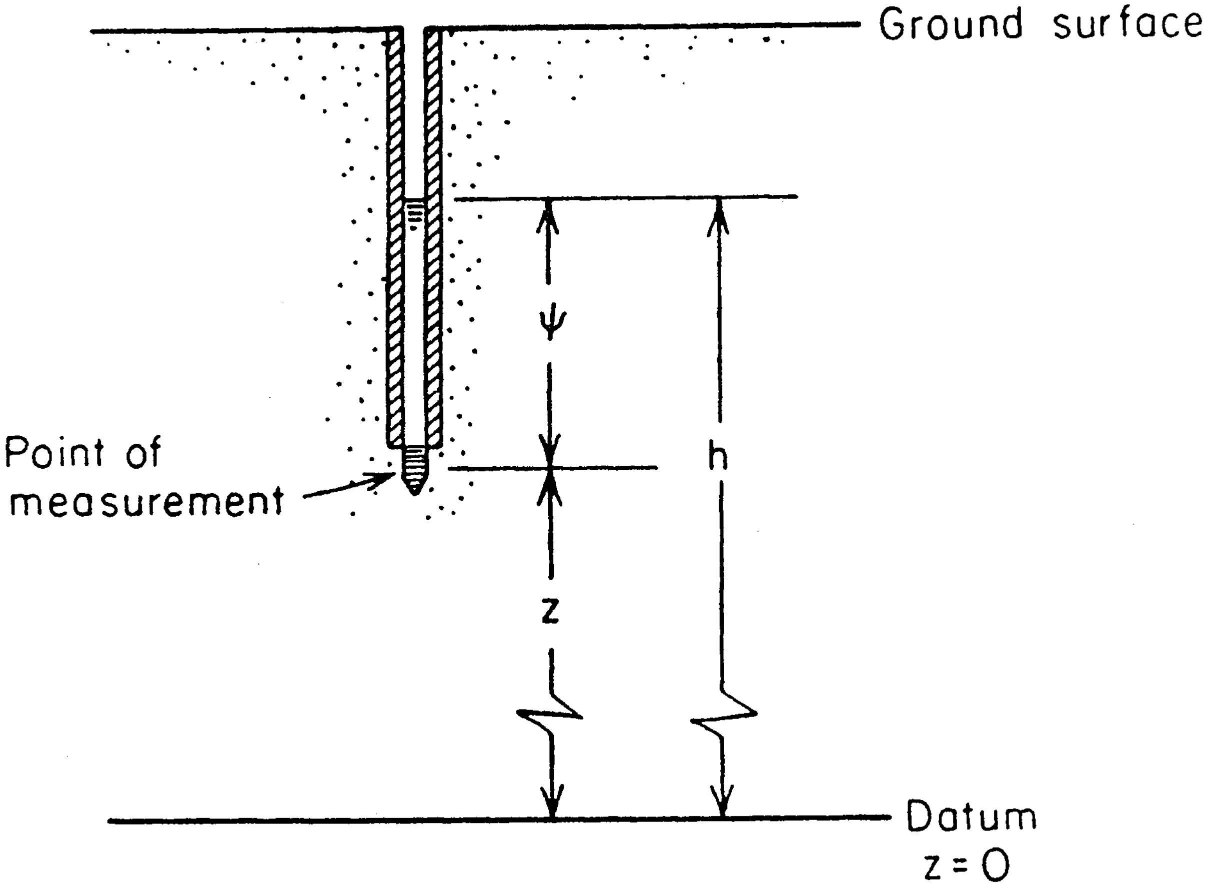

PRESSURE = P
PRESSURE CAN CAUSE WATER TO MOVE
PRESSURE = O
WATER MOVES FROM HIGH TO LOW ELEVATIONS
Z = ELEVATION
ELEVATION = 0

Figure 2



$$h = p/\gamma + z$$

$$h = \psi + z$$

h - Hydraulic head

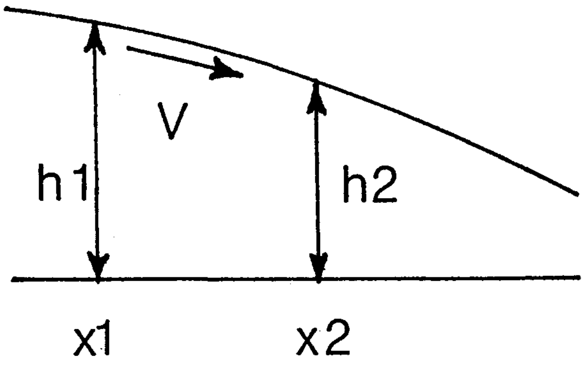
p - Fluid pressure

γ - Weight density

z - Elevation of measuring point

Figure 3

Darcy's Law



$$V = -\frac{K}{n}\frac{h2 - h1}{x2 - x1} = -\frac{K}{n}\frac{\Delta h}{\Delta x}$$

V - Average water velocity

K - Hydraulic conductivity, depends on both fluid and aquifer properties

n - Porosity of the aquifer

Water flows from areas of higher hydraulic head to areas of lower hydraulic head.

Figure 4

TYPES OF AQUIFERS

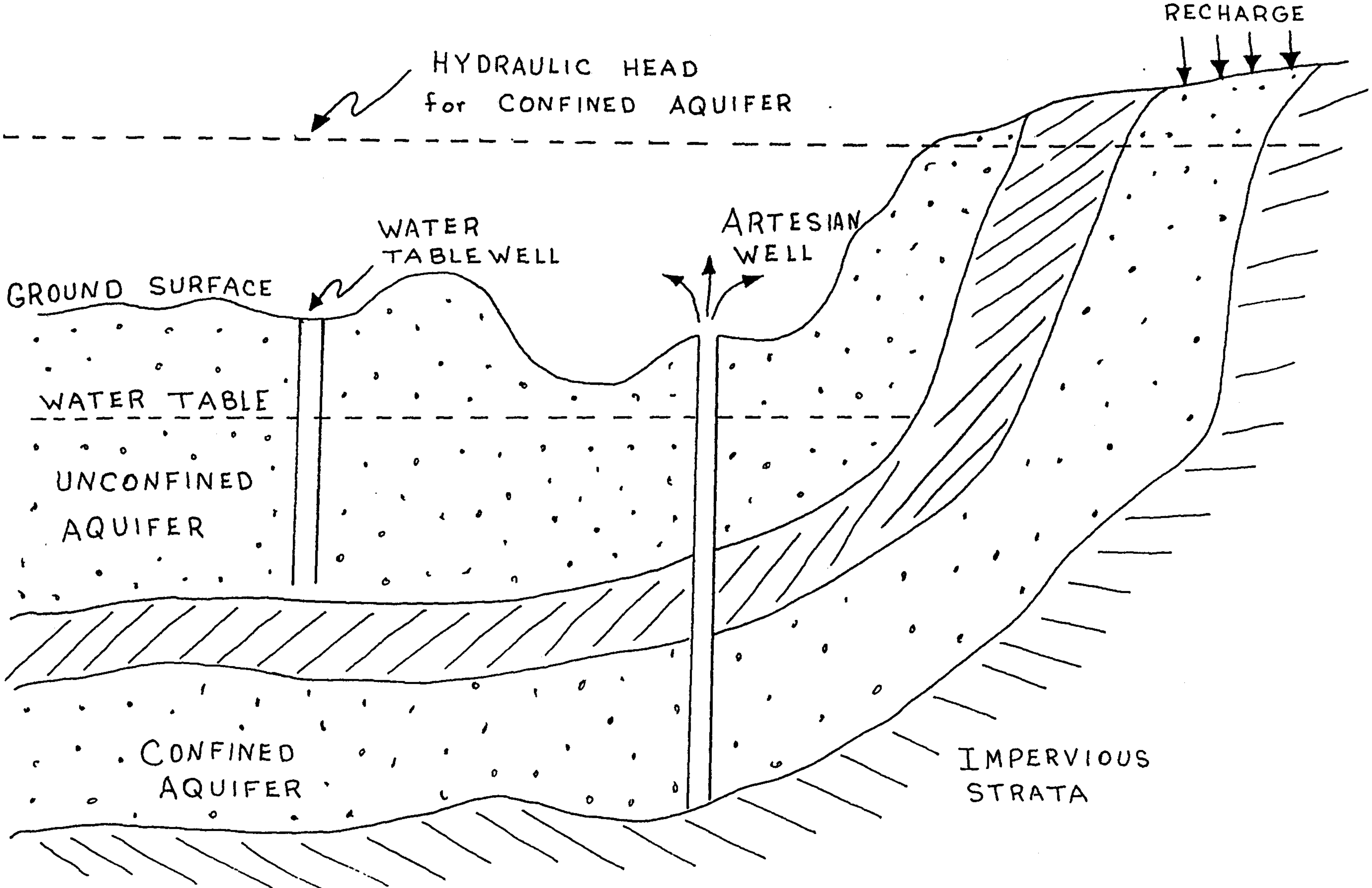


Figure 5

Divisions of subsurface water.

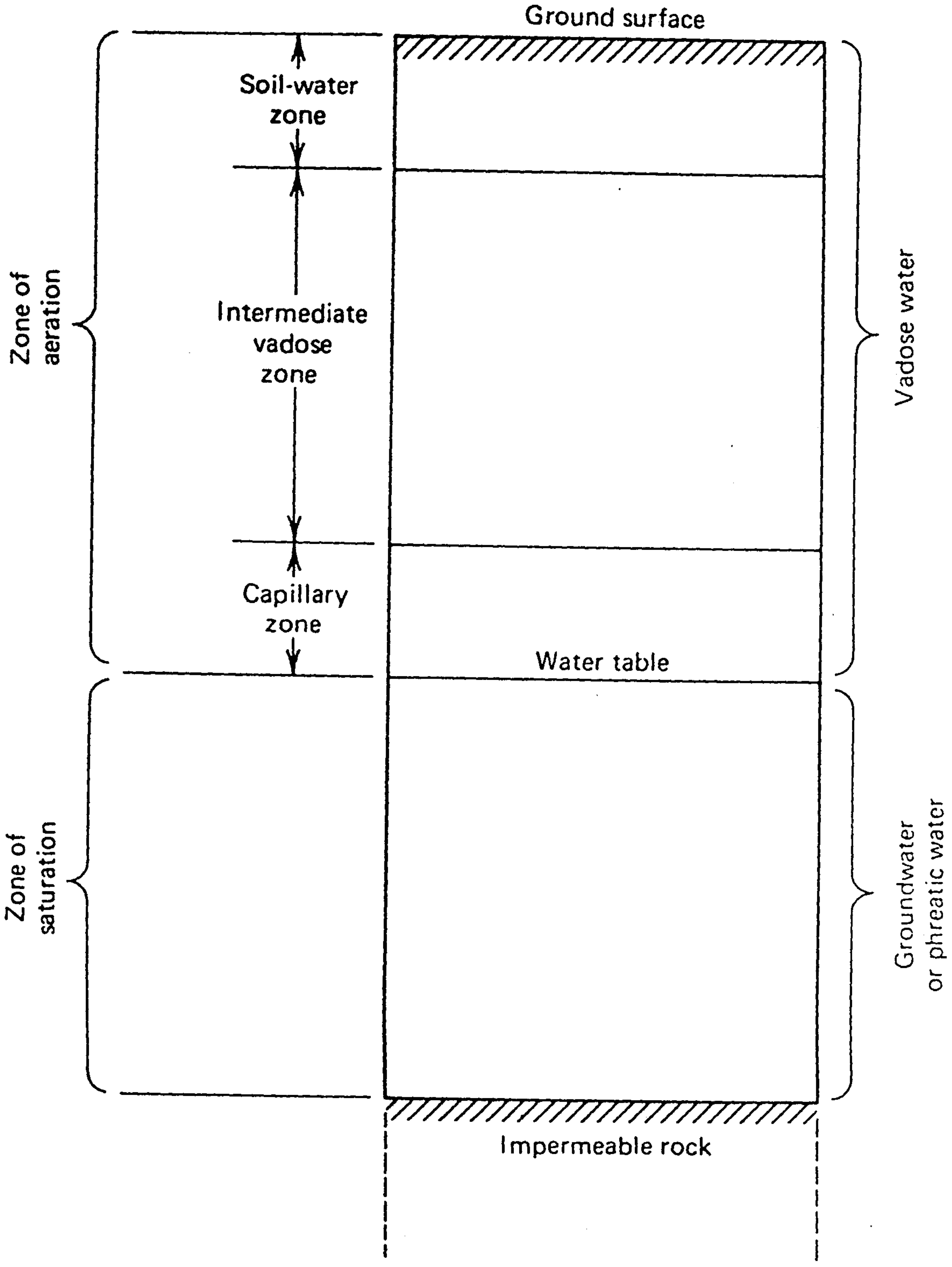


Figure 6

Kansas Geological Survey

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Hydraulic Conductivity or

Coefficient of Permeability

$$K = cd^2 \frac{\gamma}{\mu}$$

c - Dimensionless factor depending on grain shape and interconnectivity

d - Mean grain diameter

γ - Fluid weight density

μ - Fluid viscosity

Figure 8

Specific Yield (Unconfined Aquifers)

is the amount of water that can be drained by the action of gravity.

$Sy = Vw/Vt$

Sy - Specific Yield

Vw - Volume of water drained by gravity

Vt - Total volume of sample

$Sy \leqslant n$

Figure 9

Storage Coefficient (Confined Aquifers)

is the amount of water yielded by elastic properties when the hydraulic head is lowered.

$$S = \gamma b(\alpha + n\beta)$$

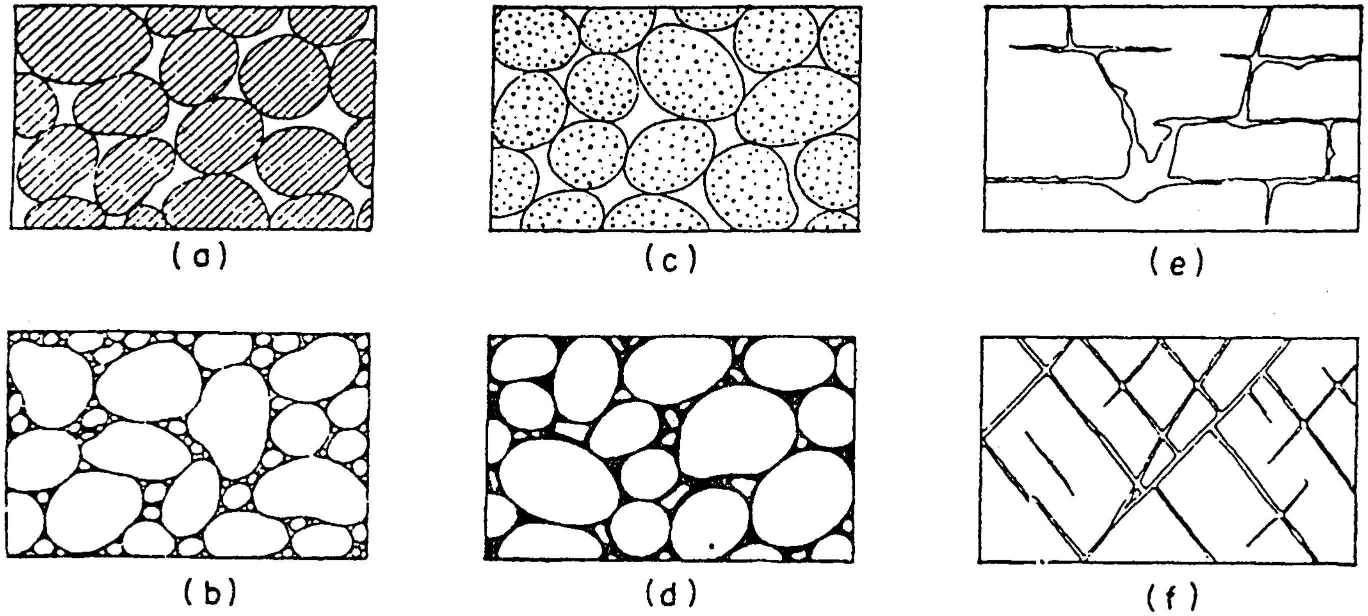
b - Thickness of aquifer

α - Compressibility of aquifer

β - Compressibility of water

Figure 10

Figure 11


(a) (c) (e)

(b) (d) (f)

Rocks	*Porosity,* %	*Specific yield,* %	*Permeability,* *gpd/sq ft*
Clay	45–55	1–10	0.001–2
Sand	35–40	10–30	100–3,000
Gravel	30–40	15–30	1,000–15,000
Sand and gravel	20–35	15–25	200–5,000
Sandstone	10–20	5–15	0.1–50
Shale	1–10	0.5–5	0.00001–0.1
Limestone	1–10	0.5–5	—

Hydraulic Head Map

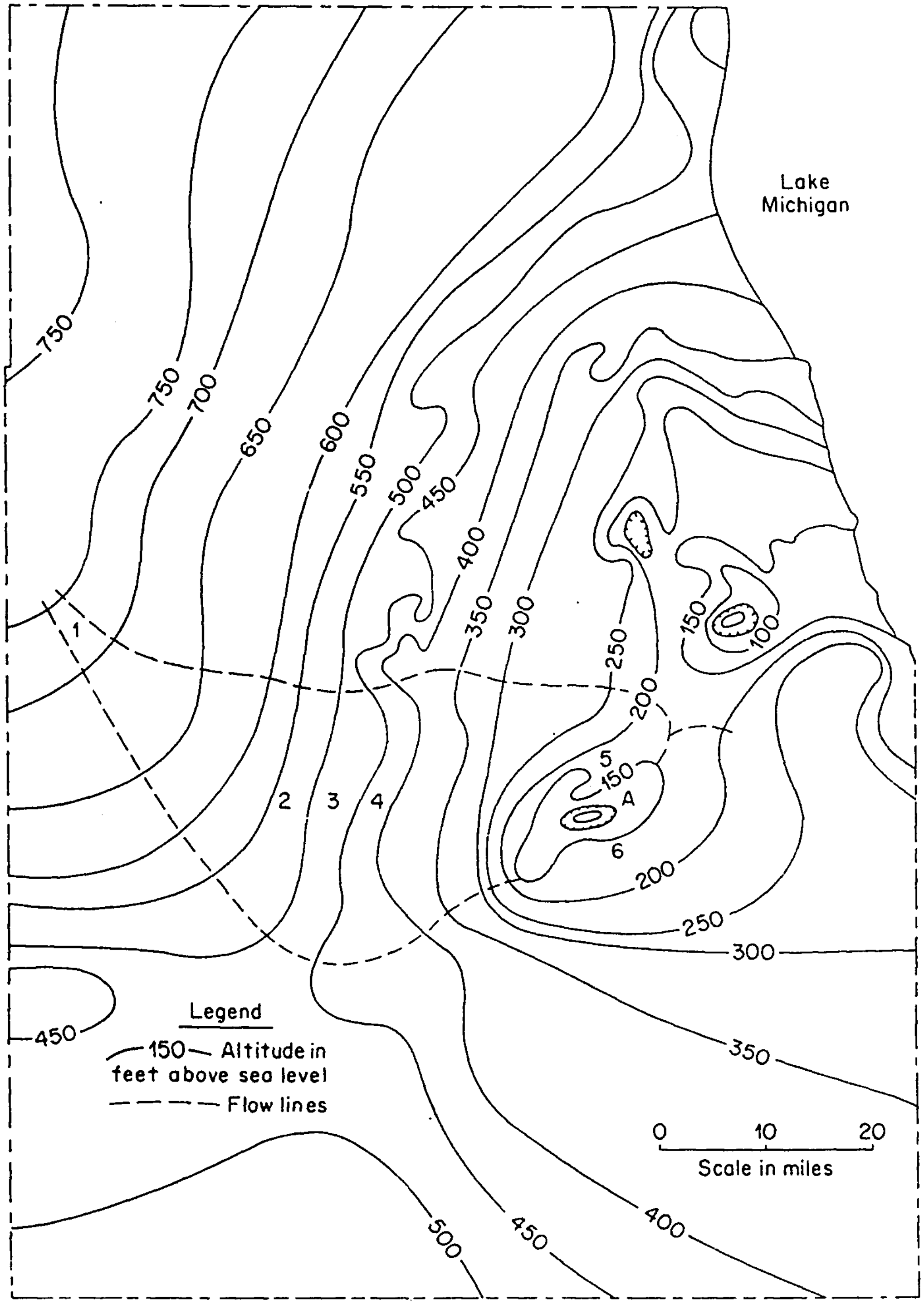


Figure 12

Kansas River Valley

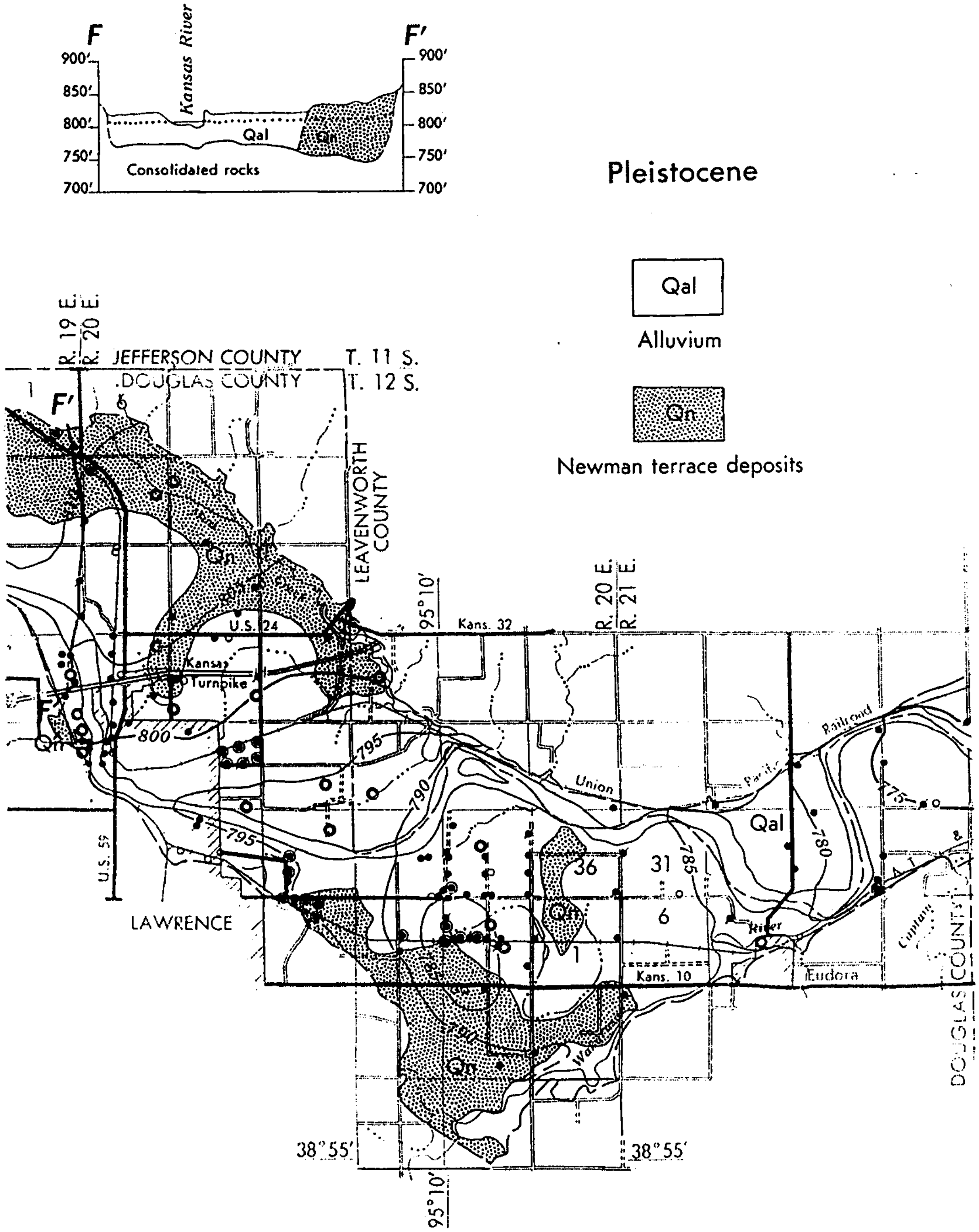


Figure 13

Average Water Velocity

$$V = -\frac{K}{n}\frac{\Delta h}{\Delta x}$$

Typical Values For Kansas

Unconfined Unconsolidated Alluvium

Ogallala and Major River Valleys

$V \approx 1\text{-}2$ft./day

Central Kansas

Lower Cretaceous Aquifer

Cheyenne and Dakota Sandstones

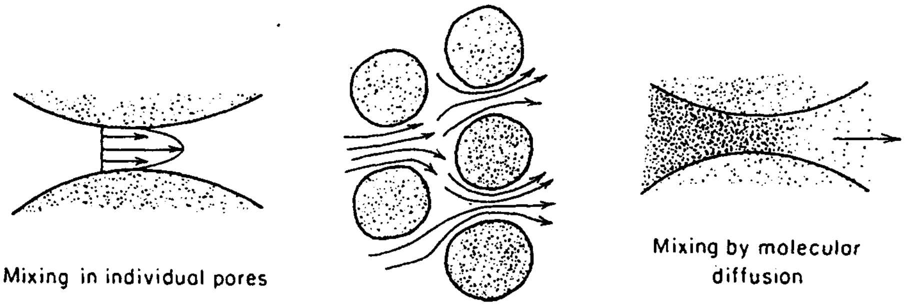
$V \approx .1\text{-}.01$ ft./day

In Typical Clays and Shales

$V \approx .01\text{-}.0001$ ft./day

Figure 14

Hydrodynamic Dispersion



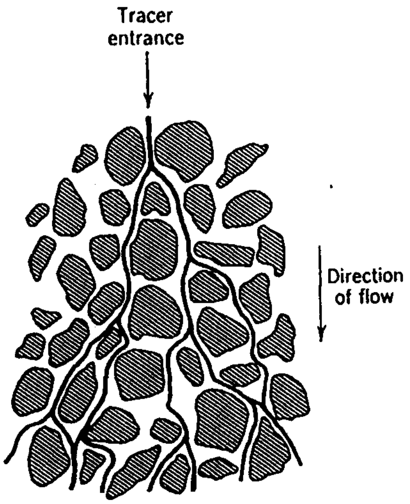
Mixing in individual pores

Mixing of pore channels

Mixing by moleculor diffusion

Figure 15

Example of Hydrodynamic Dispersion



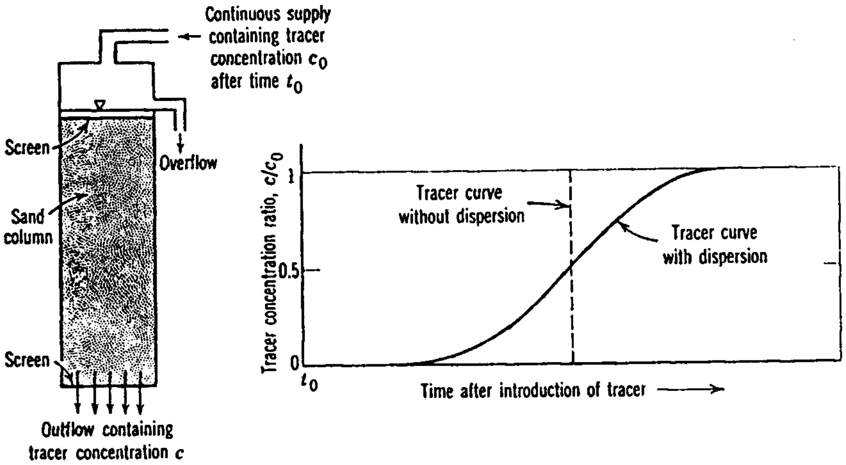


Figure 16

Distribution Coefficient

$S = KdC^{b}$ Experimentally

S- Mass of solute adsorbed or precipitated per unit mass of porous medium.

C - Solute concentration

b - Some power to be determined experimentally

Retardation Equation

$Vs/V = 1/(1 + Kd \frac{\rho}{n})$

Vs - Solute velocity

Typically $\rho/n = 4\text{-}10$

If $Kd = 1$ $Vs = V/5\text{-}V/10$

As Kd gets larger the species is more immobile.

Figure 17

DISTRIBUTION COEFFICIENTS FOR ALLUVIUM AND BENTONITE[a]

	Distribution Coefficient (mL/g)			
	Alluvium		Bentonite	
Element	Sorption	Desorption	Sorption	Desorption
U(VI)	6	60	200	170
Sb	6	80	7	50
Sb[b]	30	220		
Mo[b]	20			
I[b]	0.15	4.6		
Sr	220	180	1700	2500
Ru[b]	10	300		
Nb	1900	3500	1000	2200
Ba	3800	4000	4000	6000
Cs	8000	8000	1800	2200
Co	9000	21,000	1300	7000
Ce	>20,000	>2000	>500	>2000
Eu	>5000	>2000	>1400	>6000

[a] Data from Wolfsberg (1978) and Wolfsberg and Wanek (1982).

[b] Trace water produced by leaching test debris.

Figure 18

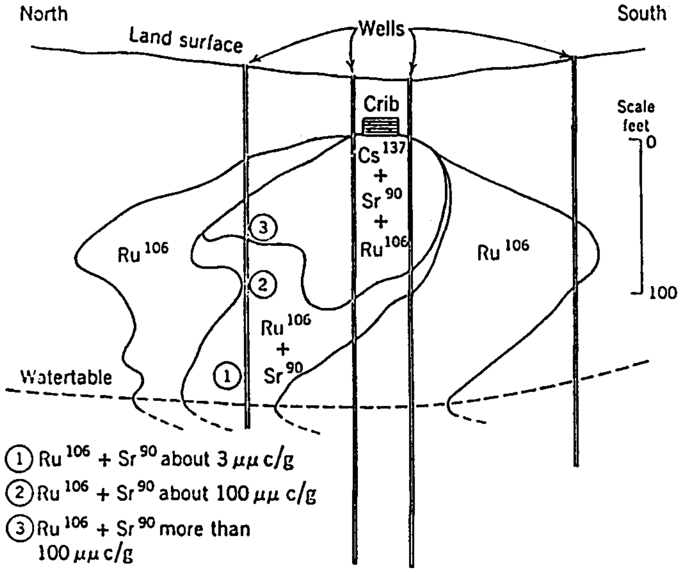


Figure 5.3 Downward movement of radionuclides from crib used for waste disposal at Hanford, Washington, January 1956. Two outer wells were drilled after use of crib was discontinued. (Diagram from Linderoth and Pearce [38].)

Figure 19