A Hydrologic Analysis of Postulated Liquid-Waste Releases Brookhaven National Laboratory Suffolk County, New York

---

GEOLOGICAL SURVEY BULLETIN 1156-E

This report concerns work done on behalf of the U.S. Atomic Energy Commission

De Laguna—ANALYSIS, LIQUID WASTE, BROOKHAVEN NATIONAL LABORATORY, SUFFOLK COUNTY, N.Y.—Geological Survey Bulletin 1156-E

A Hydrologic Analysis of Postulated Liquid-Waste Releases Brookhaven National Laboratory Suffolk County, New York

By WALLACE DE LAGUNA

STUDIES OF SITES FOR NUCLEAR ENERGY FACILITIES—
BROOKHAVEN NATIONAL LABORATORY

GEOLOGICAL SURVEY BULLETIN 1156-E

This report concerns work done on behalf of the U.S. Atomic Energy Commission

U.S. DEPARTMENT OF THE INTERIOR
March 3, 1849

UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON : 1966

UNITED STATES DEPARTMENT OF THE INTERIOR

STEWART L. UDALL, *Secretary*

GEOLOGICAL SURVEY

William T. Pecora, *Director*

**For sale by the Superintendent of Documents, U.S. Government Printing Office
Washington, D.C. 20402 - Price 25 cents (paper cover)**

PREFACE

Field and laboratory work for this report was finished in 1954. No attempt has been made to bring the material up to date; therefore, some of the statements concerning waste-management practices may not be applicable today. In addition, a great deal of laboratory and theoretical research has been done since this report was prepared on the effect of sorptive processes on the transport and dispersion of dissolved radioactivity, and on the movement of dissolved radioactivity in ground-water systems. Much work throws the light of practical experience on the theoretical and analytical studies presented here.

It is emphasized that the utmost care is taken to prevent the release of unacceptable amounts and concentrations of radioactive fluids. While the postulated releases of radioactivity analyzed in this report are not completely hypothetical, they are believed to have such a low probability of occurrence as to be virtually incredible.

CONTENTS

ILLUSTRATIONS

STUDIES OF SITES FOR NUCLEAR ENERGY FACILITIES—BROOKHAVEN NATIONAL LABORATORY

A HYDROLOGIC ANALYSIS OF POSTULATED LIQUID-WASTE RELEASES—BROOKHAVEN NATIONAL LABORATORY, SUFFOLK COUNTY, NEW YORK

By WALLACE DE LAGUNA

ABSTRACT

Application of general hydrologic principles permits an analysis of the consequences of releases of radioactive solutions. At Brookhaven National Laboratory relatively small quantities of radioactivity are available for release to the environment. Only at the reactor, the hot laboratory, the tank farm, and the waste concentrating plant is there enough activity to make an accident even a potential hazard to the water resources of the area. In these areas of interest the water table ranges from 25 to 60 feet below the land surface. The sand and gravel above the water table and below the soil zone are only partly saturated, and hence the permeability to small volumes of downward percolating fluids is lower than if the material were saturated.

About half the rainfall, 22 inches, reaches the water table. Liquid contaminants would move downward primarily by displacing water in the unsaturated zone; some mixing would take place, but some of the contaminants would be held by capillary forces in the unsaturated zone. Contaminants would not reach the water table sooner than a few weeks after release to the ground surface, and under some circumstances several years might be required for all the contaminants to reach the water table.

A leak from a buried pipeline or tank could release a larger volume of fluid before being detected, which could infiltrate to the water table through a relatively small volume of soil and cause somewhat more serious consequences than a surface release of a contaminant.

Below the water table the shape of the contaminated zone depends on the rate of lateral movement of the ground water, physical properties of the aquifer, and density differences between contaminant and ground water. Laboratory experiments with dyes demonstrate these deductions. Differences between vertical and horizontal permeability of an aquifer, due to the effects of bedding, exert a marked influence on the shape of a contaminant plume.

The average rate of ground-water movement beneath Brookhaven Laboratory is half a foot per day; for conservatism in calculating traveltime and to allow for permeability variations, a rate of 1 foot per day is used.

The effects of adsorption and ion exchange on contaminant movement are complex; extrapolations from laboratory experiments are possible only under specific circumstances. Complexities arise from the presence of several different adsorbents, different radioactive ions and different mechanisms, the presence of interfering macroconstituents in solution, and inhomogeneity of the soil. Because

of these complexities and because of the large factor of safety afforded by the slow rate of ground-water movement, laboratory experiments conducted after an accidental release, when actual compositions of waste and soil could be used, would be more informative than general laboratory experiments with idealized compositions.

Gross ion-exchange capacities of sands at Brookhaven are 200 to 300 milliequivalents per cubic foot. The exchange capacity of the Gardiners Clay is 20 times this, but ground water moves preferentially through the sand rather than the clay.

Contamination of surface waters from spills is unlikely because the soil is so permeable that contaminants will infiltrate before flowing overland any significant distance. Significant amounts of activity could reach surface streams through the sanitary sewer system, however. Traveltime from sewage plant outfall on the Peconic River to Riverhead is estimated to be on the order of a few weeks. Dilution by turbulent mixing would be important in reducing concentrations; adsorption of cations on suspended sediment is a small additional safety factor.

Effective monitoring of ground water for a suspected leak or spill depends on proper location of the well and depth of screening. This could best be done by analyzing water samples from a single fully penetrating, high-yield well so located that its cone of influence would extend outward beneath all the principal potential sources of contamination. Existing supply wells are of some value but are not fully penetrating. Monitoring surface streams, such as the Carmans or Forge Rivers, would be ineffective in detecting ground-water contamination because ground water discharging to those streams has already passed through offsite areas of use.

Remedial measures following a spill should include paving the affected area to prevent infiltrating water from driving the contaminant downward and to inhibit the spread of radioactive dust and then mapping the subsurface distribution of the contaminant. Contaminants that reach the water table could be pumped out and disposed of into a deep saline aquifer, if the costs and other complications of such measures were justified by the seriousness of the accident. Several scavenging wells might be required. Confining the contaminant in the shallow aquifer by means of an engineered cofferdam or grout curtain is not considered practicable. Alternative supplies from the Magothy Formation could be developed if necessary.

INTRODUCTION

PURPOSE

The purpose of this chapter is to describe how radioactive solutions, if they were to be released, would spread through the land and water environment—particularly in the vicinity of Brookhaven National Laboratory. This description will be largely through a discussion of general principles.

LOCATIONS AT WHICH RADIOACTIVE LIQUIDS ARE PRODUCED, USED, OR STORED

The present (1955) scope of the problem is indicated by the amounts of activity used or stored at various locations in the Brookhaven Laboratory. (See fig. 8.) Large quantities of activity can originate only in the reactor. In the reactor building the only large quantity of activity in liquid form is in the so-called "canal," a tile-lined storage chamber, in which about 60,000 gallons of demineralized water is used as a transparent radiation shield. Normally this water contains negligible amounts of activity, but during the cleaning-up operations after a slug rupture, the water has reached levels as high as 2×10^{-7} curies per cubic centimeter. This represents a total activity in the canal water of about 50 curies. In the event of serious equipment failure, the activity could be much higher.

Most experiments involving more than tracer quantities of activity, and virtually all employing curie amounts, are performed in the hot laboratory. Liquid waste from such experiments is temporarily held at the hot laboratory in two 2,000-gallon storage tanks and in several smaller tanks with a combined capacity of about 1,000 gallons. Storage for longer periods is provided at a tank farm about 750 feet north of the hot laboratory. The hot laboratory is connected to the tank farm by several pipelines. The tank farm comprises six 8,000-gallon underground tanks of stainless steel, each enclosed in a watertight concrete cell. The bottoms of these cells are 25 feet below ground. Three 100,000-gallon steel tanks for low-level waste are on the surface. An evaporating plant to concentrate the waste is at the tank farm and includes two 5,000-gallon tanks. At the start of 1954 normal operation involved waste in storage at the hot laboratory and the tank farm of about 200,000 gallons, and this waste contained a total activity of 60 to 75 curies. These quantities are relatively small because no uranium slugs from the pile are processed at Brookhaven. For this same reason the waste does not consist of the usual mixture of fission products. The waste comes from experiments for which larger or smaller quantities of particular isotopes have been specially made, so that the composition of the wastes varies considerably from time to time, and it is not possible to anticipate what might be contained in the solutions lost by a hypothetical accident.

Very much smaller amounts of activity are produced or used at several other places in the laboratory. At these places, liquid waste that may contain activity does not go directly to the sewers but goes first to holdup tanks, where it is tested before being released. If the count is above a certain very low figure, the liquid is trucked to the liquid-waste concentrating plant. The location, number, and capacity of these holdup tanks is as follows:

Location	*Number of tanks*	*Capacity of each tank (gallons)*
Chemistry	2	5, 000
Cyclotron	2	5, 000
Biology	2	5, 000
Medical	2	2, 000
Laundry (near medical)	2	2, 000
Physiology	3	275
Autopsy room	1	500

The amount of activity that the Laboratory will discharge from these tanks into the sanitary sewers is governed by the requirement that the total activity in all the liquid waste that may be deliberately discharged to the environment shall not exceed 1½ curies a year. It has been Laboratory policy, however, to limit discharge to no more than 10 percent of this amount. In practice, this means that there is seldom more than 0.001 curie in any of these smaller tanks at any one time, which is about the maximum amount of activity that could be directly discharged from one of them into the sewers. A series of accidents might result in one of the tanks having as much as 1 curie, but this would be the most that one of them would contain at one time.

Most liquid active waste, therefore, goes to the waste-concentrating plant, where it is separated into an effluent virtually free of activity and an active solid residue. The liquid is discharged to the sewers, and the solid residual, together with other solid "hot" waste (including dead rats, filter paper, and rubber gloves) is mixed with concrete, loaded in steel drums, and dumped to greater than 1,000 fathoms in the sea beyond the edge of the Continental Shelf.

The Laboratory has no burial ground for solid waste because such objects as contaminated laboratory apparatus are decontaminated as far as possible, and the remainder is disposed of at sea with the radioactive wastes. A few curies of some solid material containing insoluble radioactive substance with a short half life are buried, but leaching by rainwater can hardly pick up enough activity from such material to be of interest. Therefore, only at the reactor, the hot laboratory, the tank farm, and the concentrating plant are there enough radioactive materials to make an accident even a potential hazard to the water resources of this area.

GENERAL NATURE OF AQUEOUS TRANSPORT OF RADIOACTIVITY

In any discussion of the environmental release of radioactive solutions, the most important questions will be the direction and rate of movement of the contamination, the change with time of the boundaries of the area involved, and the distribution of the concentration of activity. Each of these items depends on a wide variety of factors, and much more is known about the operation of some of these factors than about others.

The following discussion is concerned first with movement down through the vadose zone to the water table. In the vadose zone the sand and gravel are moist but not saturated, and the controlling forces are due to surface tension. A distinction will be attempted between the results of a sudden spill, which might release a large volume of liquid in a relatively short time, and a leak, which might release the liquid slowly over a much longer period.

A second distinct stage, in which surface tension is no longer a factor, is begun when the escaping liquid reaches the water table and moves laterally with the ground water through a saturated aquifer. Such movement is more uniform and is for that reason better understood by hydrologists, although the presence of two liquids—an aqeous chemical solution and the surrounding native water—introduces complications.

Important and probably governing factors during the movement both above and below the water table are adsorption and ion exchange. Unfortunately, this report is not able to assess the effects of these processes, not only because they are physical-chemical phenomena lying outside the area of competence of the geologist or the hydrologist, but also because even the scientists most familiar with the problem find them complex. No proper evaluation of their influence could, therefore, be included in the following discussion, and such quantitative estimates as are attempted of necessity neglect these factors. In every case, however, the effects of adsorption and ion exchange will be to slow down the rates of movement, to restrict the distances traveled, and to reduce the concentrations of activity in the liquid phase. The omitted aspects of the problem, therefore, are safety factors and ensure that rates of movement and concentrations discussed in this report are overestimated.

After describing the probable patterns of movement, an attempt will be made to analyze the problem of monitoring the ground water and surface water for the purpose of detecting contamination if it occurred or was suspected. The conclusion of this section may be anticipated by stating here that in this writer's opinion no currently possible program of monitoring ground water appears to have much merit and that a program of monitoring streamflow, while presenting fewer fundamental difficulties, would apparently require far more effort than it would be worth.

Possible remedial measures to be attempted in the event of serious contamination of the ground water will be discussed. Although such contamination would create difficult problems, there appear to be several ways either of holding the contamination in place, of transferring it to a deeper unused aquifer or, as a last resort, of developing alternative water supplies.

The general conclusion is that hazardous contamination of the ground water or surface water is most unlikely even in the event of a serious accident, although to be positively certain would require a more thorough investigation of some of the general aspects of the problem.

MOVEMENT IN THE ZONE OF AERATION

NATURAL MOISTURE DISTRIBUTION IN ZONE OF AERATION

From the bottom of the canal in the reactor building or from the ground surface in the waste-storage area, the depth to the water table is about 50 or 60 feet. From the bottom of the buried tanks at the waste-storage area, the depth is about 25 feet. The general figure of 50 feet will hold for most of the currently utilized areas in the Laboratory tract; although under some of the little used eastern parts of the Laboratory grounds, the depth to the water table is only a few feet.

Below the 2 to 4 feet of surface soil, the sand and gravel in the intermediate zone between the water table and the soil contain about 10 percent of water by volume. Where the depth to the water table is 50 feet, roughly 5 feet of water is contained between the land surface and the water table. This water is held as very thin layers on the surface of sand grains, in thin rings around the points where grains touch one another, and in the smallest of the openings between grains. In such positions surface tension resists the force of gravity, and the water is held in the sand above the zone of saturation.

The 5 feet of water is not evenly distributed through the vadose zone. Above the water table is a narrow fringe, in which the pores are largely saturated by water raised by capillary forces. This capillary fringe has no sharp upper boundary; at progerssively higher levels the openings that are filled with water become progressively smaller. Above the capillary fringe the moisture content decreases rapidly at first and then more slowly, although there are no sharp discontinuities. In the Pleistocene sand underlying the Laboratory, the moisture content at any point more than a few feet above the water table is determined not by the height above the water table but by the size distribution of the pore spaces. The finer textured beds, with a higher proportion of small pores, contain more moisture—captured from water descending from the surface.

Even well above the water table the little rings of water that are retained around the contact points of sand grains, and the minute irregular droplets held in the smallest of the pore spaces, are generally in contact with one another. This is shown by the electrical conductivity of the material, which is low but measurable. Movement in the liquid phase is, therefore, possible but is slow, and the capillary conductivity (effective permeability) of a sand that has drained by gravity for a few weeks is low. The final equilibrium water content (specific retention) of a sand layer 50 feet thick is approached but never reached on Long Island. A small increment of water added to the top of such a layer almost at equilibrium would move downward slowly. A larger increment, by filling more and larger pore spaces, would increase the capillary conductivity, and so would move down more rapidly. Only under special circumstances could enough water ever infiltrate through the relatively fine surface soil to saturate the sand below, so that the permeability determined for saturated material would almost never be reached.

NATURAL MOISTURE MOVEMENT IN ZONE OF AERATION

During an average year the precipitation is evenly distributed and amounts to nearly 4 inches a month (about 45 in. per yr.). Recharge to the water table is not evenly distributed, however, for during the warm weather from May to October evapotranspiration equals or perhaps exceeds precipitation, and little rain gets past the soil zone. In fact, during most summers a moisture deficiency occurs in the top few feet of soil; that is, the moisture content is reduced below the limit reached by natural drainage. With cooler weather, evapotranspiration is greatly reduced, the soil moisture deficiency is made up, and water from the surface starts moving down toward the water table. If heavy rains at this time supply sufficient quantities of water to this wave of reacharge, the water will move down rapidly. The first of the water may take several weeks or, in a dry year even months to reach the water table at a depth of 50 feet, and the increments from individual storms will have blended. With the arrival of warm weather, the infiltration of additional water through the surface ceases; however, slow movement and drainage below the surface apparently continue all summer, although the rate of arrival of water at the water table is greatly reduced. Actually, the pattern in any year is determined by the weather of that year. In some summers an unusually heavy rainstorm may soak the soil, and part of this moisture may start on its way toward the water table. Heavy rains associated with the very infrequent late summer or fall hurricanes may be responsible for an appreciable wave of recharge. On the average, however, about half the annual precipitation of 45 inches, or about 22 inches, makes its way to the water table. In some of the following discussions an average annual recharge of 2 feet will be used to facilitate generalized computations.

EFFECT OF ADDITIONAL INFLUX OF A "SLUG" OF WATER

A small increment, or "slug," of fluid added suddenly to the surface of the ground in the summer or early fall, when the upper few feet of soil has been dried below its specific retention, will be held near the surface. The amount that can be so retained will depend on the moisture deficiency at that time. The average maximum is estimated to be 4 inches of water, but the amount may be less. The moisture deficiency is largely created in the top 1 or 2 feet, which is directly affected by evapotranspiration. Some transpiration losses extend down as far as 10 feet.

The horizontal spread of an increment of liquid, say a spilled drum of contaminated liquid, is governed by the shape and texture of the surface of the ground. Ponding, as well as variations in the permeability of the surface, may cause much more infiltration in some places than in others.

In theory, at least, at any point where the amount of infiltrating water exceeds the moisture deficiency, some of the infiltrating liquid will reach the upper edge of the interconnected disseminated water in the vadose zone and bring about a slow general movement of liquid down to the water table. In the cooler months, when natural recharge is active, any increment at the surface will immediately start on its journey to the water table.

The infiltration of enough fluid through the surface of the ground to start a general movement in the vadose zone toward the water table does not mean necessarily that the same fluid will reach the water table. Adsorption and ion exchange will remove and hold back some, much, or even all the dissolved substances, such as contaminants. Neglecting, however, these important factors, the 5 feet of water retained in the vadose zone still must be considered. Water added at the top will in part displace bodily downward the water already present in the vadose zone and in part mix with it, but the relative effectiveness of the two processes is not clear. If the vadose zone were invaded from above by contaminated liquid, the descending wave of liquid, as it entered pore after pore, would release and mix with the small quantities of water held in these pores by capillary forces. The mixing is not by turbulence but by diffusion, which in such small spaces acts quickly. As the now diluted liquid moved downward, some of it would be left behind to mix in turn with the liquid arriving from above. In the pores below, the advancing front of contaminated liquid would be further repeatedly diluted, until a certain thickness of the advancing front had been transformed into nearly pure water. A simple experiment has served to confirm this inference. Water colored with indigo carmine, a dye which is little absorbed on sand, was poured onto the top of a glass column filled with sand, which had been saturated with water and then well drained. As the colored water moved down, a screen of apparently pure water developed ahead of it; and by a judicious choice of the quantities involved, blue water poured in at the top could force out clear water at the bottom. Undeniably, however, there was some mixing.

The exact amount of mixing under these circumstances, although of theoretical interest, would probably not be a critical consideration in the event of a real spill. If the amount of spilled liquid penetrating per square foot of surface were a small but still appreciable fraction of the moisture held between the surface and the water table, a small proportion of the fluid would reach the water table; but by far the greater part of the contaminated fluid would be held by capillary forces in the zone of aeration. To illustrate, assume that 1 foot of contaminated liquid has infiltrated into the ground in an area in which the water table is at a depth of 50 feet, and assume that mixing is inoperative. The second assumption was apparently made by Wind (1951), a Dutch hydrologist. He introduced a fluorescein dye solution into one end of a column packed with damp sand to force out the moisture contained in the sand and attempted to determine the moisture content of the sand by measuring the volume of clear water that emerged ahead of the dye. The 5 feet of water held in the vadose zone, if we accept the concept of pistonlike displacement, will be displaced bodily downward so that the contaminated water will occupy the top 10 feet of sand below the surface, and 1 foot of water will pass through the water table into the main body of ground water. If this happened in the summer or early fall, further movement would be small until natural recharge began in the next winter. If the 2 feet of pure water added at this time also is assumed not to mix with the water already in the ground, by the following summer the top 20 feet of soil and sand would contain pure water, and the contaminated water would lie between 20 and 30 feet below the surface. The next cycle of natural recharge would carry the water down so that it would lie between 40 and 50 feet below the surface, and not until the third winter after the postulated accident would the contaminant reach the saturated zone and start to move laterally.

For comparison, assume that the 1 foot of infiltrating contaminated water mixes completely with the 5 feet of water in the ground before any recharge to the water table takes place. After this mixing, one-sixth of the mixture would move down past the water table and carry with it one-sixth of the contamination. The next winter's natural recharge of 2 feet, if assumed first to mix with the remaining five-sixth, would then carry down with it about one-quarter of the contamination; but just over one-half of the original spill would still remain in the vadose zone. Each succeeding winter would in turn bring down two-sevenths of the remainder.

A possibly more realistic intermediate example would be 1 foot of infiltrating contaminated water mixing sufficiently with the water already in the ground so that the lower limit of the contamination would eventually come to rest about 15 or 20 feet below the surface. The following winter's recharge then would drive small amounts of fluid down past the water table, but the greater part would not reach the main body of ground water until two or three winters after the accident.

The degree of mixing in a homogeneous medium does not, however, appear to be the determining factor in establishing the rate or pattern of downward percolation that would develop after a sudden introduction of contaminants. Infiltration through the surface would be unevenly distributed and might well be concentrated in one or a few spots. The sand and gravel below the surface do not have uniform vertical permeability, so that streamers of liquid would advance well ahead of the main body and might even reach the water table. Under any system of mixing, however, only a small part of the introduced fluid would reach the water table quickly. A large part of the activity would be retained in the vadose zone. Clearly the natural recharge during subsequent winters would be a major factor in flushing the retained contamination down to the water table.

In contrast to a spill, in which the contaminated liquid would be all released in a short time, is a leak, in which the escaping fluid would be released over a long period involving perhaps years. Let us imagine such a circumstance in connection with a buried tank or pipeline, for a long continued undetected leak at the surface of the ground is very improbable. The most important difference between a spill and a leak of long duration from a buried tank or pipeline is that, at the site of the leak, a relatively large volume of escaping liquid might pass down through a comparatively small horizontal section—perhaps hundreds of cubic feet through a single square foot of sand. This cross section would possibly increase somewhat with depth because, in addition to the vertical force of gravity, a lateral force of surface tension due to the moisture gradient is operative. As the liquid moved downward from the point of escape, some of it would be attracted toward the drier sand at the sides of the column. The downward flow of liquid would be largely confined to the central column, however, because the moisture content of the outer zone would be only slightly increased and this zone would still have a low capillary conductivity.

It appears certain, however, that a definite volume of liquid lost slowly by a small leak will move down through a smaller cross section than the same volume spilled on the ground, and if this is true, the liquid may well reach the water table sooner and leave a smaller proportion of its volume trapped in the pores in the vadose zone. In addition, the liquid would come in contact with a smaller volume of soil and so would lose less activity by adsorption; and also it would form a narrower ribbon of contamination in the ground water. Where equal volume of liquid and equal amounts of contamination are involved, therefore, a slow leak could have more serious consequences than a sudden spill.

The amount of dilution to be expected in the vadose zone is difficult to estimate, but it appears to depend in part on the depth of contaminated liquid (cubic feet per square foot of surface) infiltrating into the ground. If an inch or so of liquid infiltrated past the surface, dilution would probably be significant; but if several feet of liquid made their way past the surface, dilution could be negligible.

The rate of movement from the surface to the water table depends on the degree of saturation of the sand below the soil, and this also is governed largely by the amount of water infiltrating past the surface. Where water is ponded on the surface and the soil and sand below receive all the water they can pass, the water will reach the water table at a depth of 50 feet within a few hours or in a day or two, as shown by observations at the time of a pumping test on well S6434. Numerous but less specific observations of the times at which ground-water levels start to rise in the winter or spring show that several weeks or months may be required for the natural recharge to reach the water table at depths of 50 to 100 feet. The velocities clearly vary over a wide range and are a function of both the amount of water added at the top, the vertical permeability, and the vertical distance through which the moisture travels.

ARRIVAL AT THE WATER TABLE

The shape of the contaminated volume that would be formed at and below the water table by the arrival from above of contaminated liquid depends on the rate of arrival of this liquid at the water table. The wide range of velocities possible in the vadose zone make it difficult to consider more than a few simple general or limiting examples.

As a first example, consider what happens to the 2 cubic feet of water contributed annually to the water table by a representative single square foot of ground surface. In the Brookhaven Laboratory area most of this recharge arrives at the water table during the first 6 months of the year. As the porosity of the upper Pleistocene sand is about one-third, 2 cubic feet of water will fill the voids in about 6 cubic feet of sand. The average rate of movement of the ground water is a half a foot per day. As the fresh increment from above arrives at the water table, it will be entrained by the moving ground water and, as a first approximation, formed into a ribbon a foot wide, 90 feet long, and 0.07 foot thick.

A large spill would probably arrive at the top of the saturated zone over a period of time that would vary with seasonal and geological factors. The arriving fluid would spread out roughly radially around the point of inflow at the top of the zone of saturation in an ideal isotropic medium and would be entrained by the moving ground water

799-427 O—66——3

into a body having, again as a first approximation, a semicircular cross section and a length depending on the period over which the fluid entered the ground water. As the average rate of movement of ground water at Brookhaven is about half a foot per day, the length of the water body in feet would be approximately half the number of days required for the spilled fluid to arrive at the water table. The cross section, semicircular in an ideal isotropic meduim, in square feet would be equal to approximately six times the amount in cubic feet introduced in 1 day (the quantity introduced in 1 day divided by a porosity of one-third and divided by a length of half a foot).

In the actual environment the sediments are bedded, the vertical permeability is less than the horizontal permeability, and the cross section would probably be a flattened semicircle or semiellipse with the long axis at the water table.

MOVEMENT IN THE ZONE OF SATURATION

LAMINAR FLOW OF GROUND WATER

Except in such aquifers as boulder beds or cavernous limestones or basalts, ground water never moves fast enough in nature to produce turbulence; and in the Upton area, the motion of the ground water does not approach turbulence. The water molecules, therefore, follow flow lines that are virtually fixed, except as the rise and fall of the water table may slightly alter their pattern. These flow lines, moreover, do not cross or intersect; and although two adjacent lines may separate and pass on opposite sides of an obstacle, once the obstacle is passed they will again assume their former relative positions. Only where additional water infiltrates from above, or is added in some other way, can a flow line come between two previously adjacent flow lines. Flow lines can be forced closer together or spread apart, however, as a passageway narrows or widens. When flow lines are forced closer together, the flow is faster; and the converse also is true. The shape of a bundle of flow lines also can be altered, as the shape of a channel changes. Adjacent flow lines, however, remain adjacent.

The water in adjacent flow lines may, however, move at different rates, so that the molecules in any flow line can be displaced longitudinally with respect to those in neighboring lines. As they move through any passageway, the molecules at the center move most rapidly, while those near the walls are retarded by the friction of viscosity; indeed the molecules touching the walls are sometimes regarded as fixed. The branching and rejoining of the irregularly sized channels through which the water moves prevent any flow lines from long remaining in the central position of most rapid movement. By separating adjacent bundles of flow lines and routing them around opposite sides of obstructions, these reticulations are themselves probably an important mechanism for causing a reduction in the extremes of velocity such as are observed in a straight capillary tube. Natural aquifers have many random constrictions and widenings of passageways, with a consequent complex pressure gradient distribution and pattern of movement on a near-microscopic scale.

The difference in rate of movement along adjacent flow lines would lead to some reduction of concentration of contamination in the direction of flow, but this process acts in conjunction with diffusion. The thermal agitation of the molecules of water—and of solute if contamination is present—enables them to move from one flow line to another. The distance through which molecules may move by diffusion is proportional to the square root of the time involved, so that, although diffusion is incapable of causing mixing over distances of many feet, it is quickly effective over the minute distances involved in the pores of a saturated sand.

The results of a simple experiment showing the general configuration to be expected are illustrated in figure 1. A watertight wood box, 20 cm deep, was equipped with a plate-glass bottom 50×100 cm, and so arranged that water would flow through it lengthwise. The box was then filled with clean medium sand, and a hypodermic needle was carefully introduced from above until its beveled point just touched the glass. A small quantity of dye solution, slightly diluted with alcohol so that its specific gravity very closely approximately that of the water, was then slowly injected so that it formed a circular spot 2 or 3 cm in diameter. The lateral rate of movement of the water in the box was about 45 cm per hr, a velocity much too slow to cause turbulence but many times the natural ground-water velocity in sandy media such as those in the Upton area. As the dye spot moved forward, it increased in length by 1 cm for each 4 cm of average advance, as measured to the center of the spot. The front and back ends of the spot became progressively less sharp as it lengthened, apparently owing to mixing and dilution, and after the spot had roughly tripled or quadrupled in length, no further elongation could be measured. The spot probably continued to elongate, however, and the failure to detect this was due to the difficulty of observing the dye where it was too greatly diluted. A proper study would involve a quantitative determination of concentrations along the axis of the spot and a more sophisticated method of expressing its length—possibly in terms of a standard deviation. The most important single question that might be determined is whether the lengthening of the spot and therefore the dilution are directly proportional to the distance traveled, as suggested by the data shown in figure 1, or whether they bear some other relation to the distance. They also may be a

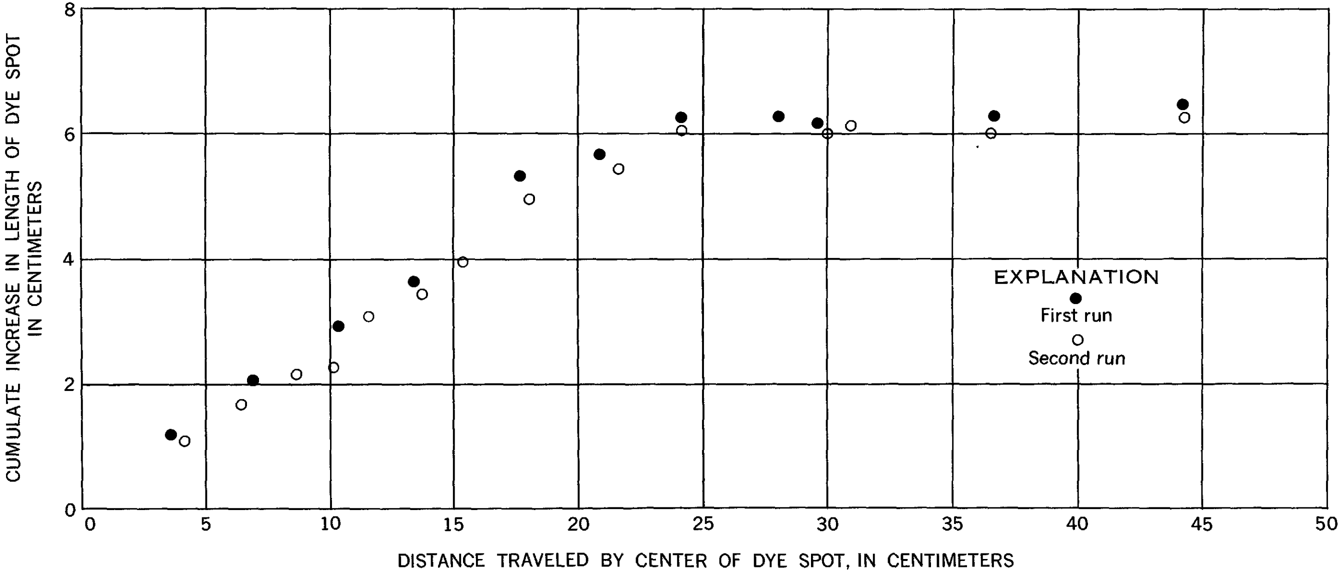

FIGURE 1.—Lengthening of dye spot due to hydrodynamic process.

function of the velocity used. Although the dye, indigo carmine, used in this experiment is not noticeably absorbed by the sand, some adsorption probably takes place, which would affect the observed results. In fact, with other dyes, all the lengthening and dilution observed could be produced by adsorption, although in this experiment independent observations showed that the observed results were due to purely hydrologic factors.

The processes described above operate only to produce elongation in the direction of movement. The writer was for some time convinced that a bundle of flow lines, split and rejoining as they passed around sand grains, would gradually become separated, so that the spot described in figure 1 would get wider as well as longer. This process was called "scattering," but later discussions with C. V. Theis of the U.S. Geological Survey have convinced the writer that the only mechanism that can cause a widening of a dye spot while moving through a homogeneous medium is diffusion. The photograph shown in figure 2 was made looking up at the glass plate set in the bottom of the same box used for the measurement of spot lengthening. In this experiment the dye-alcohol-water mixture is being injected at two points at a uniform rate. The length of each dye stream is about 90 cm, and the traveltime is about 2 hours. The small amount of widening observed can be explained by diffusion and by slight residual density differences. "Scattering" does not exist, and the difference between the rate of lengthening of the dye spot and its very slow rate of widening is a measure of the greater effectiveness of the forces related to the flow of the liquid as compared to molecular diffusion.

IMPORTANCE OF DENSITY DIFFERENCES

Density differences, however, may be capable of greatly speeding up and aiding the effectiveness of diffusion to increase the cross section of a single body or continuous stream of contaminated liquid moving in and with the ground water. If the foreign solution is more dense than the ground water, it will settle through the ground water, and the two will, therefore, move in slightly different directions. This slight crosscurrent acts in much the same way as a difference in the rate of movement along adjacent flow lines because it will bring new liquid filaments adjacent to one another. This effect does not occur on the same intimate scale as that produced by the lengthening of the dye spots, for the liquid bodies involved are much larger than single pore spaces. As mixing and dilution take place along the boundary between the two fluids, liquid of intermediate density is formed, an additional factor which appears to be of considerable importance. This mixture will then settle less rapidly than the un-

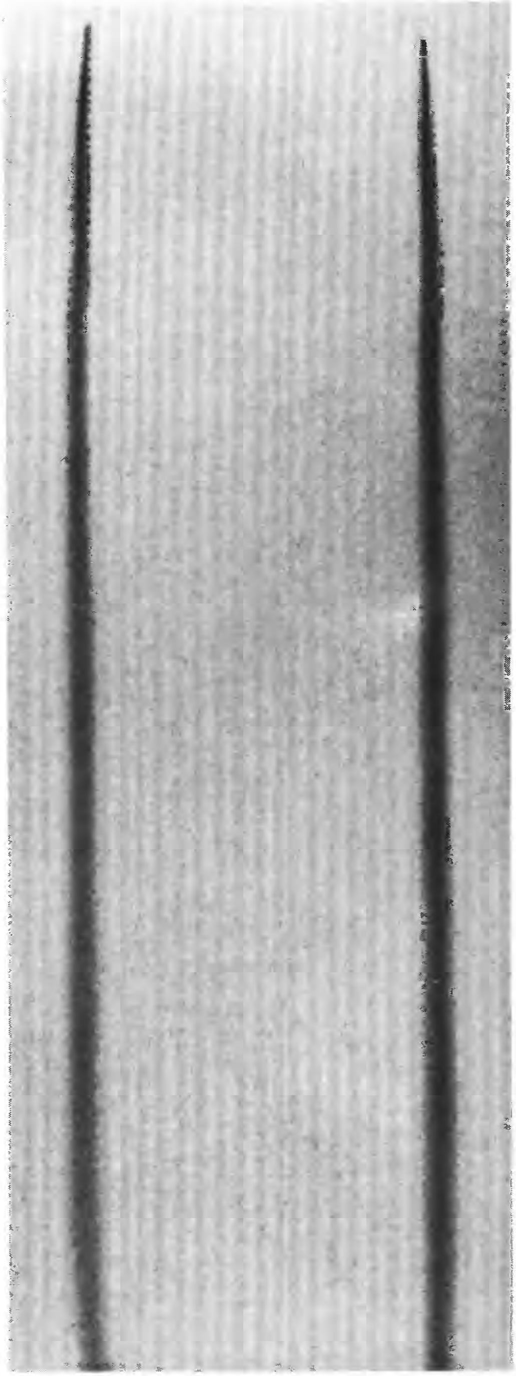

FIGURE 2.—Failure of dye streams to get wider suggests fallacy of "scattering."

diluted solution, which forms the core of the settling mass. Thus a disruptive tendency of considerable potential is created by the initial dilution; its method of operation is complex.

A comparatively simple example would be afforded by an environment in which the ground water itself was stationary. This can be illustrated experimentally by the arrangement shown in figure 3, in which the three beakers contain sand saturated with water. In each beaker a small quantity of dye was introduced into the saturated sand by a pipette pushed down along the beaker wall, so that a dye spot was formed on the inside against the glass. The outline of the spot was then carefully drawn on the outside of the beakers with a wax crayon and the beakers set aside. The photograph shows the result after 2¾ hours. The spot on the left was made with a dye solution having a density of 0.9996 g per ml; the tap water had a density of 0.9960 g per ml. (These densities were determined by Dr. R. W. Stoenner of the Brookhaven National Laboratory staff.) The density of the dye solution in the middle beaker was adjusted by adding alcohol to 0.9956 g per ml, as close as was feasible to the density of the tap water. An excess of alcohol, added to the dye in the right-hand beaker, produced a density of 0.9920 g per ml. The movement of the dye spots in the outer beakers is obvious.

The front edge of the spots is dark and sharp. Some mixing took place at the back edges of these spots, but all the dye has moved down or up out of some of the area the dye originally occupied. The unchanged size and position of the middle spot shows the negligible movement resulting from diffusion alone under these circumstances.

In the left-hand beaker the denser dye sank through the sand, displacing the less dense water below it. The displaced water, moving out and up and around behind the spot, filled in the pores in the area vacated by the spot. The pattern of movement resembles convection. The driving force was the density difference between the water and the dye (0.9996−0.9960=0.0036), or about 3½ parts per thousand. This density difference, however, must not only move the dye down, it must also move an equivalent volume of water up; so, to compare the density difference with gradients produced by a pressure difference it must be divided by two, giving about 1.8 parts per thousand. Natural ground-water gradients in the Brookhaven area are about 1 part per thousand.

As the dye spot moves through the sand, it is elongated by the hydrodynamic process already mentioned. The elongation and dilution of the dye spot (fig. 1), which was entrained and carried laterally by slowly moving water, were similar at the front and rear edges. Where density difference is the driving force, however, as is shown in

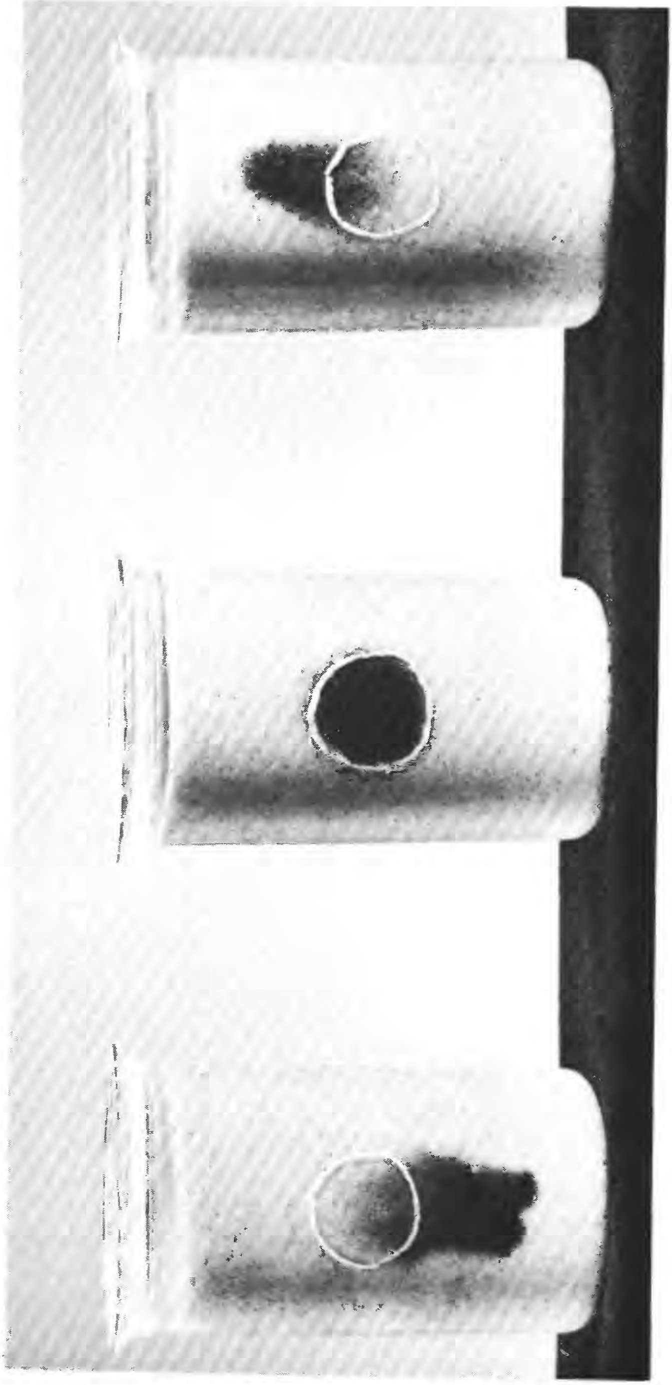

FIGURE 3.—Movement of dye spots in sand due to density differences.

figure 3, the result is not symetrical. Any dye solution at the rear edge of the spot, which is diluted even slightly, is then acted on by a reduced vertical force and descends in consequence less rapidly, pulling away from the main mass. Or at least the diluted solution attempts to pull away, for as the main body of the dye spot descends, the void behind it must be filled. At the front edge of the spot any filament of dye that gets ahead of the main body and is diluted, even slightly, slows down in consequence of the resulting diminution of density difference and is overtaken by the main body of the spot coming down behind it. This mechanism alone would tend to make the front edge of the spot sharp and the rear edge of the spot diffuse, but it is aided by the pattern of movement along the flow lines. The water displaced at the front of the sinking spot moves up past the edges of the spot, and the resulting difference in rate of movement along adjacent flow lines increases the rate of the mixing along the front and sides of the spot. The diluted dye so formed is, however, carried up and around to the rear of the spot, where it fills the pore spaces vacated as the main body of dye moves down.

Even a relatively dilute solution will be acted on by vertical forces of a magnitude comparable with the nearly horizontal force represented by most natural water-table gradients. Foreign solutions will, therefore, settle down through the natural ground water as both move forward, followed perhaps by a diffuse and elongate tail, which will reach up and back from the sinking central mass of the denser solution.

The discussion of the movement of contamination below the water table has so far been based on an assumed ideal homogeneous aquifer. In a real aquifer, where the vertical permeability varies with depth owing to the bedding, the mass of active fluid will widen as it sinks. The widening takes place at the contact between a more permeable bed above and a less permeable bed below, the descending fluid piling up on the lower bed and then spreading out laterally. Change in the cross sectional area of the contaminated fluid may not be marked, but the change in shape of the cross section into a thinner and wider ribbon may increase the opportunity for other mixing mechanisms. Unpublished data of the Nassau County Department of Public Works on the movement of waste chromate solutions dumped into pits in Nassau County suggest that these heavy solutions sink until they reach a thin bed of silt or fine sand, then they flow along the surface of this bed, widening laterally as they move.

The foregoing discussion has considered “slugs” or “spills.” A slow leak over a long period of time might raise some additional problems. The contaminant would form a continuous ribbon in the ground water, and its elongation in the direction of movement would be unimportant, inasmuch as it would be mixing with itself. If the ribbon were denser so that it settled through the ground water, the resulting disruption and mixing would be effective in producing dilution. Some of the elements of the problem are shown in figure 4. This photograph was taken looking at the side of the same sand-filled box used in some of the previous experiments. Clear water is flowing from right to left at a rate of approximately 1 cm per min. Two fine tubes feed dye solution continuously against the inner surface of the glass side of the box. The lower dye stream is a mixture having very nearly the same density as the water; the upper dye stream has a density of 1.1, and so it sinks. The lower stream maintains its position and shape and is obviously being diluted slowly. The upper stream, just as obviously, is being disrupted and diluted. As with the sinking spot, the lower edge of the sinking dye stream is dark and sharp. Mixing and dilution may be taking place along the sides, but the diluted dye is dispersed above the main dye stream, in part, because it is less dense than the main dye stream and so sinks less rapidly.

The lower dye stream, moving horizontally, appears to hit the descending upper stream and mix with it. Actually, the lower dye stream passes behind the upper stream, for flow cannot move in two directions at once through the same pore spaces. The volume occupied by the descending dye stream is as impenetrable to the horizontally moving dye stream as a solid obstruction and forces the less dense water to flow around it. This is responsible for some of the observed dilution, but it has other results as well. The horizontally moving clear water, obstructed by the descending dye, must develop an additional head to flow around it, and this local increase in gradient affects the dye stream so that it descends at a smaller angle. This combination of countermoving liquids is unstable, and, as shown in figure 5, the water below may force itself up past or through the barrier. At any place where the trapped water starts to break through, the barrier is thinned and weakened, so that more water is able to move past in this area. In figure 5, the water is forcing itself between the dye stream (5 percent cupric chloride) and the glass, and the stream remains continuous. A wider sheet in descending would be more likely to be actually penetrated or even disrupted by the ground water.

The effect of bedding on a descending dye stream is shown in figure 6. The upper dye stream has the same density as the ground water; the lower is more dense. As much dye is being fed into the lower stream as into the upper, but the lower stream is being forced away

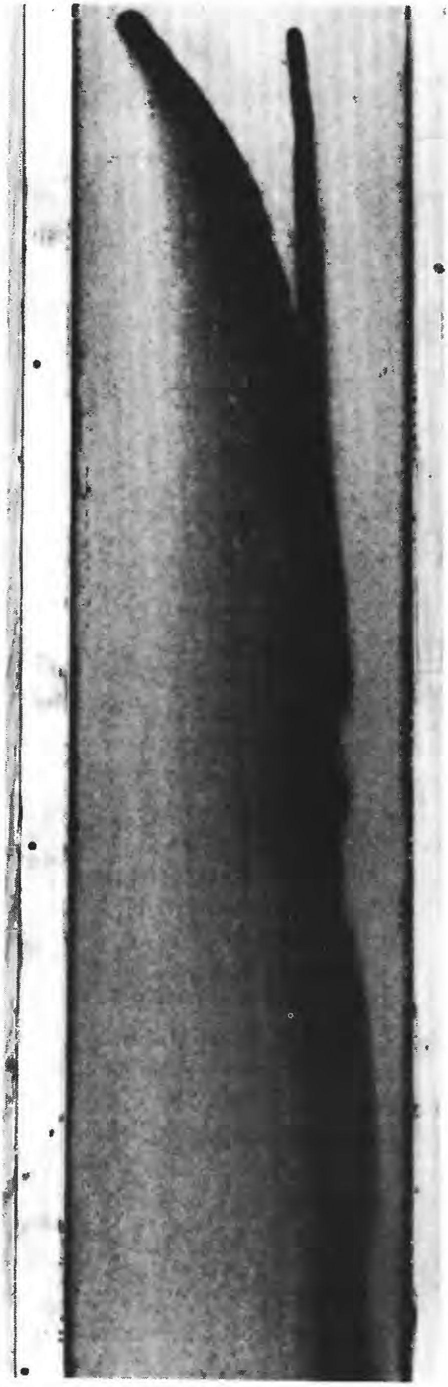

FIGURE 4.—Movement of dye streams in simulated moving water.

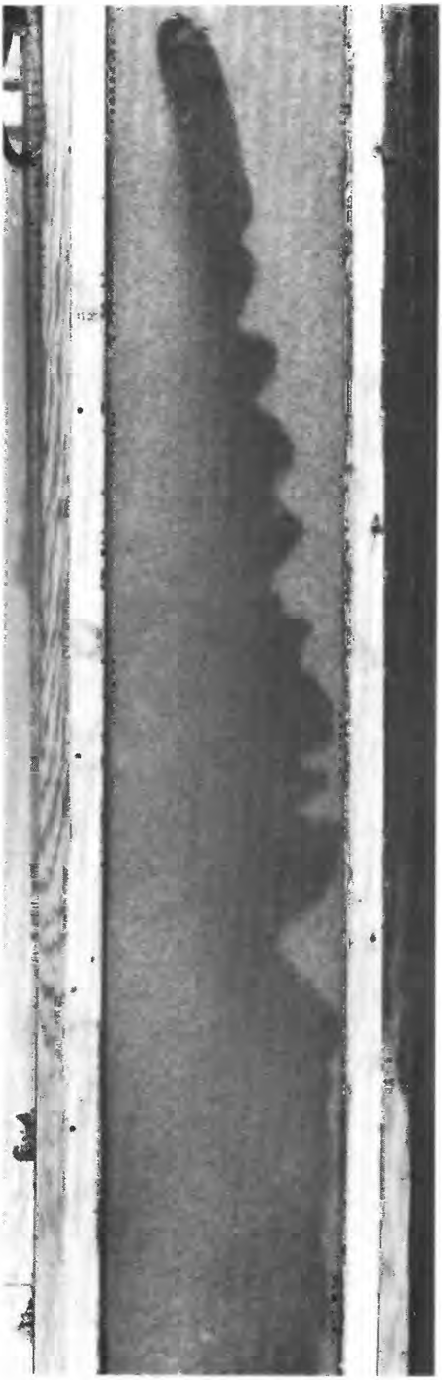

FIGURE 5.—Descending dye streamer blocking moving ground water.

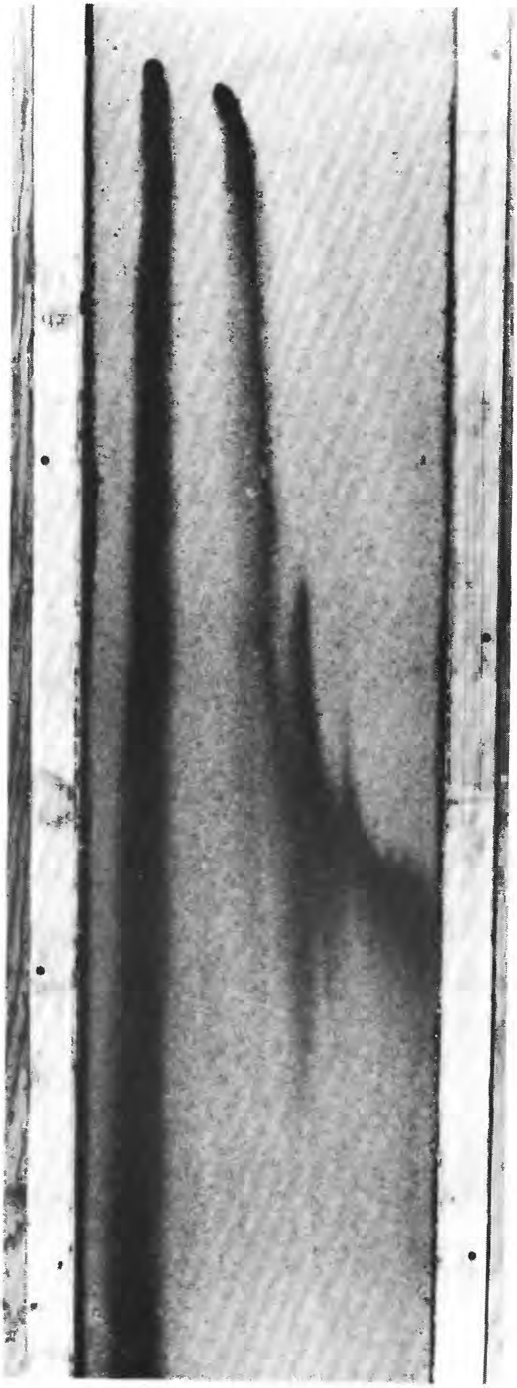

FIGURE 6.—Effect of bedding on descending dye stream.

from the glass by the obstructing effect described in the preceding paragraph. As it sinks, however, the lower dye stream comes against slightly less permeable layers and spreads out. These wide places show up in figure 6. A section through the sandbox a few inches to the rear probably would show a pattern for the descending dye stream much more like that in figure 4.

RATE OF MOVEMENT OF THE GROUND WATER AND AN INCLUDED CONTAMINANT

The average speed of movement of the ground water in the upper Pleistocene sand in the Brookhaven Laboratory area is about half a foot per day. Although the systematic difference is not marked, the lower part of the aquifer appears to be less permeable than the upper part, and unquestionably local variations in permeability are caused by comparatively fine- or coarse-grained beds tens or perhaps even hundreds of feet long. It would be wrong to regard the average speed of half a foot per day as the maximum rate of advance of the ground water. The rate of movement of the ground water, determined from hydrologic tests, is an average rate for all the molecules and, at any instant, some of the molecules are traveling faster and some slower than the average. Any single droplet of a foreign fluid can, however, hardly maintain a faster than average rate of movement over a considerable distance in passing through the reticulations of the many pores of a normal sandy sediment. Because of differences in permeability of the different layers, a rate of 1 foot per day will be used in the following discussion of the possible rate of movement of contaminants in the ground water rather than the average rate of half a foot per day.

Because of this relatively slow rate of movement, isotopes with a half life of less than a year introduced into the zone of saturation would have little chance of reaching the boundary of the main Laboratory tract except in much-reduced concentrations, even if large quantities were spilled, and isotopes with a half life of less than a few months hardly can be regarded as a ground-water hazard even within the Laboratory grounds. This can be seen from the following table, which lists the eight isotopes most dangerous if ingested (National Bureau of Standards, 1953), their half lives, and the distances in which their concentration would be reduced by decay to 1 percent of the original.

For comparison, the distance from the waste-storage tank farm to the nearest of the Laboratory's supply wells is about 2,000 feet and to the boundary of the Laboratory tract is more than 1 mile.

Effect of decay alone, with rate of movement of 1 foot per day

[Few of the isotopes listed will actually move underground at the same rate as the ground water. They will be slowed down by adsorption and ion exchange, so much so that even in the relatively clean sands underlying Brookhaven, many of them will be virtually stagnant. Natural decay will therefore have a longer time to act than the table suggests]

Isotope	*Half life*	*Distance to reduce activity to 1 percent*
Bismuth-210	5 days	33 ft
Yttrium-91	57 days	380 ft
Zirconium-95	65 days	430 ft
Calcium-45	180 days	¼ mile
Cerium-144	275 days	⅓ mile
Iron-55	4 yr	2 miles
Promethium-147	4 yr	2 miles
Strontium-90	25 yr	11 miles

ADSORPTION AND ION EXCHANGE

The importance as well as the complexity of the somewhat similar mechanisms of adsorption and ion exchange stem from their ability to selectively remove molecules or ions from solution. Most laboratory experiments, and in consequence much of the information derived from them, involve systems that have only a single solute and a single adsorbent. Some experimentation has used a solution containing several solutes, but hardly any work has been done with mixtures of adsorbents because of the complexity of interpreting the results. Radioactive solutions in the environment constitute, however, a complex mixture of ions and adsorbents, so that a prediction of results using either a theoretical approach based on simple experiments or an empirical approach based on attempts to duplicate the actual field conditions, would be unsatisfactory. The difficulties involved have been summarized specifically for inclusion in this report by Henry C. Thomas while on the staff of the Department of Chemistry of Yale University.

THE MOTION OF FISSION FRAGMENTS THROUGH SOIL

By Henry C. Thomas

When one examines the multiplicity of problems involved in any attempt to predict the detailed behavior of the motion of radioactivity through beds of soil it speedily becomes evident that quantitative extrapolations from laboratory experiments will be nearly if not entirely worthless unless these experiments have been devised to apply to a specific actual situation. This is by no means to say that all knowledge derived from somewhat idealized laboratory experiments is useless. Qualitative understanding of the phenomena to be expected will be essential, and this knowledge can be derived only from quantitative laboratory work. The following discussion shows why detailed predictions are generally too much to expect.

In any small-scale laboratory experiments in which chromatographic studies are made on beds of soil, one would first either make every attempt to render the bed homogeneous or he would deliberately try to reproduce in a column the natural composition profile of the soils in question. Because the latter project would necessitate a most judicious scaling down, many attendant uncertainties would arise, particularly in the matter of handling the flow rates. In any project, experiments with inhomogeneous beds would be of much greater complexity, both in performance and in interpretation, than would experiments on homogeneous beds.

With a homogeneous column, the assumption could be made that there was only a single adsorbent of arbitrary character. One could not, however, assume that the adsorbing processes involved any single and specific type of adsorption. For example, a soil conglomerate might contain appreciable amounts of hydrous ferric oxide in addition to clay minerals. The clay minerals will function at least partially through ion exchange, and the hydrous ferric oxide will bring about adsorption without the reintroduction into the solution of equivalent amounts of ions. Generally, one would have to assume an arbitrary adsorption isotherm with no simplifications owing to knowledge of specific mechanisms. The experiments would reduce to efforts to determine this isotherm.

Since the isotherm is arbitrary in nature, one would have no advance knowledge of how to take into account the interaction on the adsorbent of the microamounts of fission fragments of interest with the macroamounts of the other constituents of the solutions. This difficulty may, however, prove to be of no consequence. If the macroconstituents of the solution do not react chemically to change the character of the adsorbent, it may be shown that the nature of the macroconstituent is frequently a matter of indifference. Thus, the behavior of trace cesium on a clay mineral is not significantly dependent on whether the supporting electrolyte is sodium or potassium. More complex situations would have to be investigated, at least in a preliminary fashion to establish this independence.

It is immediately evident that if the character of the adsorption processes in the soil bed is changed by macrochemical reaction with the solution, then the nature of the isotherm at a given level in the column will depend on the previous history of the solutions. A case in point would be an adsorption on ferric oxide and its dependence on the pH of the solution. In such a situation the "isotherm" determined would depend on the height of the column. There might be little resemblance between results obtained with a column of given length and those obtained with a column twice as long. This would be regarded as a trivial and removable complication in the laboratory, but would be considered as more serious in connection with an actual spill.

One can suppose, however, that these difficulties will disappear, even in case of a real accidental spill. Unfortunately, another difficulty just as formidable as the others remains. In case of a spill of fission fragments dissolved in a nonreacting solution, the analog in the laboratory is a column fed with a solution of definite and constant concentration. In case of a spill of a reactive solution, perhaps one containing much free acid, the initial adsorption processes will serve to give some complicated partial separation of the fission fragments involved. Parts of the soil fed with the chemically spent solution will then be fed with variable and largely unknown concentrations of the radioactive species of interest. The corresponding chromatography problem in the laboratory becomes much more complex and, without knowledge of the isotherm, it is impossible to solve if the input concentrations are entirely unknown.

Let us look at the character of the laboratory jobs in which the important but obvious difficulty of inhomogeneity of the bed has been removed. The following cases, arranged in order of increasing complexity, are indicative of the nature of the problems involved. The category to which a given spill belongs would always be determined on the basis of its particular characteristics.

1. A column fed with a solution in a nonreacting solvent containing a definite concentration of a single radioactive species in trace amount.

   This is the simplest case of elution chromatography. The deduction of the arbitrary isotherm is a relatively simple matter. Certainly any general program of study would start with experiments would be needed to map out even one case on a single every type of soil bed that comes into question as well as for every fission fragment of importance. Although the enormous number of experiments that would be needed to catalog all the possibilities presents a formidable undertaking, such a program would be the most fruitful method of study.
2. The same problem but with a mixture of fission fragments all in noninterfering trace amounts.

   This situation can be constructed from the results of experiments envisaged in case 1. In addition, a sufficient number of experiments must be done to demonstrate the mutual noninterference of the radioactive species.
3. The same problem with the radioactive species fed into the column in varying but known concentrations.

If the various species do not interfere, this case can be handled on the basis of the results of case 1.

4. A column fed with a solution in a nonreacting solvent containing a pair of components that mutually interfere on the adsorbent.

   It seems improbable that this situation could arise with any synthetic radioactive species. Nontrace amounts of the naturally occurring activities would, however, afford a case in point.

   The chromatography problem immediately becomes immensely more complex with mutual interaction. If the character of the interaction were not known, the results would in all probability defy accurate interpretation. A very large number of experiments would be needed to map out even one case on a single adsorbent. The results could be summarized in empirical two-solute isotherms, but any prediction from such results applied to a case not previously examined would be extremely hazardous.

5. The same problem as case 4 with variable input concentrations.

   In a two-solute case of this character, some of the basic mathematics has been examined on the supposition that the mixed adsorption isotherm is known. For any but the simplest conceivable isotherm the partial differential equation which is obtained can be handled only through numerical methods. To this writer's knowledge, no attempt has ever been made to examine this case in the laboratory. It is possible, even in the two-solute case for known isotherms, that the unavoidable experimental uncertainties, such as channeling in the columns, would render the work largely valueless.

6. In the above discussion, we have always implied that the chromatographic experiments have been carried out under equilibrium conditions. In any real experiment nonequilibrium effects will always be present. If these effects are large, the results of the experiments are obscured and the interpretation becomes more difficult. Little or no nonequilibrium work has been done on two-solute chromatography. This is, however, a difficulty which need give little concern. The motion of water in the soil is so slow that the equilibrium approximation will always be near to the true state of affairs. Except possibly for the effect of downstream diffusion, all nonequilibrium effects can safely be neglected. It should be remembered, however, that qualitatively all nonequilibrium effects have the same type of influence on a concentration profile in a chromatographic bed. The mere presence of a diffuse front is no evidence of downstream diffusion, although the diffuse front may well be empirically described as due to diffusion.

Even if the complexities indicated above did not make practically impossible a sufficiently inclusive experimental program to map in advance all the possibilities in accidental spills, such a program would not be justifiable because of the simple circumstance of the slow motion of the ground water. In nearly all cases of accident in which fission fragments should get into the ground stream, there would be ample time to devise and carry out appropriate laboratory work which would afford, at worst, educated guesses on the future course of the activity.

Because specific data for an interesting accidental release are not available, detailed computations of the effect of downstream diffusion will not be undertaken. The possible magnitude of the effect in various accidents is apparent from an examination of the curves given by Lapidus and Amundson (1952). Depending on the parameters involved (flow rate, diffusion constant, capacity of adsorbent, and fractional free volume of bed), one finds for a column of adsorbent of given length that, while a sharp breakthrough (no diffusion) will occur at volume 1250, in an unfavorable case the activity level may rise to 1 percent of its initial value at volume 800. In soils of low capacity, however, such a large effect would unlikely be observed. Nevertheless, in situations where very low activity levels must be considered, the possible effect of downstream diffusion must not be neglected. It would be very much worthwhile to undertake a fairly extensive computation program and tabulate in usable form the various possibilities of this effect.

ANTICIPATED INFLUENCE OF ADSORPTION

The conclusion to be reached from Prof. Henry C. Thomas' work, and one that was amplified somewhat in conversation with him, is that any specific quantitative predictions as to the influence of adsorption and ion exchange on the movement of activity in the soil will have to wait until after the event. A study of the pattern of movement during the early stages follow a spill could then be used to predict the future rate of movement of the contamination and the concentrations of activity to be expected. Also, as the composition of the spill would then be known, laboratory experimentation could be directed at a specific problem.

This acceptance of the necessity for a delay in determining the quantitative effects of adsorption does not mean that remedial measures would be hampered in the event of an accident. The movement of liquids in the zone of aeration is slow and depends in no small part on infiltration of moisture from precipitation, unless the volume of the spill should be large. Also, the movement of contamination below the water table is slow, so that an accident, no matter how serious, does not appear to be the type of event in which the effectiveness of remedial measures will depend on prompt action. Attempts should be made, however, to determine in advance what the effects of adsorption and ion exchange would be because this information would be essential for any realistic evaluation of the potential danger of contamination of the water resources of the area and would therefore affect matters of policy, which are influenced by the nature of the danger. Such matters of policy include the use, storage, and disposal of radioactive liquids, particularly radioactive wastes. They might also include policy on the monitoring of water resources in the area by suggesting the most appropriate methods of analysis; however, the problem of getting adequate samples of surface water and ground water is so serious as to raise doubts as to the value of any water-monitoring program.

Basically the effect of adsorption is to temporarily remove material from solution at a point where the concentration of that material in solution is increasing. As the liquid moves on, if more dilute solution reaches the point of interest, the material will be returned to the solution from the adsorbent. The observed effect at any point in the line of travel is a delay in the arrival of activity. Figure 7 shows the results obtained when a small volume of a solution of sodium chloride and fluorescein was added to water flowing through a column of sand 5 feet long. Samples of the water emerging from the column were taken every 7½ minutes and analyzed both for chloride and fluorescein. As can be seen in the diagram, the chloride first appeared after 52½ minutes; the fluorescein took just twice as long to appear. In fact, the chloride had nearly all passed through the column before the first of the fluorescein showed up in the effluent; with a slightly longer column, the separation would have been complete. Obviously, there was much more adsorption of the large molecules of the organic dye fluorescein than there was of the chloride ions. How much, if at all, the chloride was delayed is not known. The rates of movement of the solution in this experiment were much more rapid than those of natural ground water. This experiment points out the difficulty of determining the rate of movement of ground water with a tracer, for any adsorption or ion exchange will affect the apparent rate of movement of the tracer.

Another possible effect of adsorption is to stretch out the length of a single body or mass of contaminated liquid in the direction of motion. In fact, it is not possible to be sure how much of the lengthening of the dye spot, shown in figure 1, was due to hydrodynamic processes and how much to adsorption. Another example is shown in figure 7 in which the fluorescein dye continued to come through for

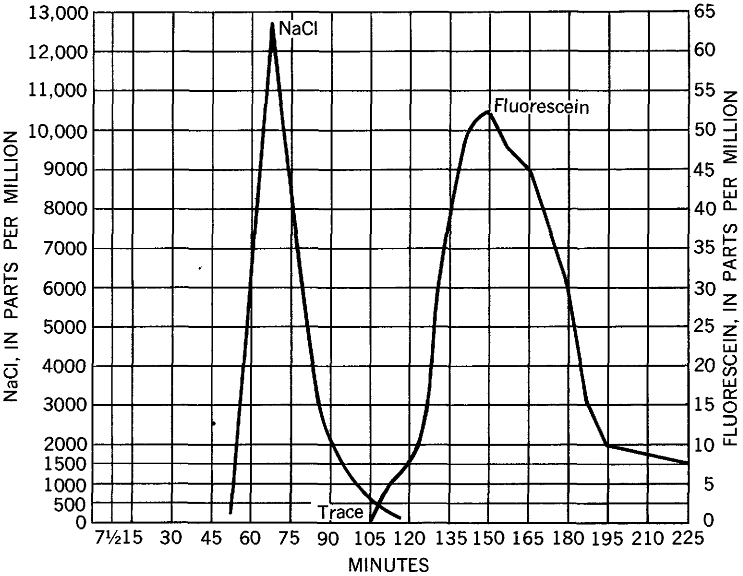

FIGURE 7.—Separation of NaCl and fluorescein by differential adsorption on sand.

some time after the peak concentration had passed and continued for some time; in fact, after the last sample of effluent had been collected. Adsorption, because it operates only in the direction of travel, may not be effective in diluting the long, thin streamer of contamination that would result from a steady leak. Even the rate of advance of the front end of a long thin streamer would be slowed down, however; and the resulting delay, where adsorption was marked, might well have much the same practical result as permanent removal.

Figure 7 also illustrates another effect of adsorption: its ability to separate materials in solution. The chloride in this example has been separated almost completely from the fluorescein. The same mechanism is, of course, the basis for chromatographic separation. Because of differences in adsorption, therefore, all the radioactive contaminants would not travel at the same rate through the soil or through the sand aquifers. Even an imperfect knowledge of their relative speeds may suggest the contaminants most likely to be the first to arrive at a particular point and so indicate both the most appropriate analytical procedures and the level of activity that would create a hazard, as both of these depend on the particular element or isotope that is migrating. In general, anions are less adsorbed in the soil than cations, and, as a consequence, anions are freer to travel through the soil. Radioiodine is also apparently little adsorbed (Watkins, 1954). Of the longer lived isotopes, ruthenium, with a half life of about 1 year, appears to be the most rapid migrant under field conditions.

A related problem is the rate of entry of contaminated ground water into a pumped well. This will be largely determined by the shape of the front of the advancing wave of contamination. This shape, in turn, will depend in part on the way in which contaminants might be introduced, whether by a slowly developing leak or a sudden spill, but it will depend also on adsorption and perhaps on hydrodynamic effects. Prof. Henry C. Thomas' opinion was that in general the contamination would appear slowly in a sampling well, in part, because the first arrival would be the isotope least affected by adsorption.

The mineral composition, and in particular the minerals composing the silt and clay fractions, are given by Faust (1963) for several representative formation samples, together with values for the base-exchange capacities for these samples. The values indicate the amount of cation removed from a relatively concentrated solution under the conditions of the experiment and are, of course, not a quantitative measure of the amount of cations that would be removed from dilute solutions containing only trace amounts of active cations. The data are, however, indicative of the relative ion-exchange capacities of the main stratigraphic units. Although compared with many aquifers that contain more clay, the relatively clean sands at Brookhaven have a low ion exchange capacity; nevertheless every cubic foot of this sand has a base-exchange capacity of 200 to 300 milliequivalents. In traveling even a short distance, therefore, a contaminated solution would come into intimate contact with large quantities of adsorbent material, and very substantial delay and retention of the contaminants could be anticipated. Even the upper Pleistocene deposits, therefore, have a tremendous potential adsorptive capacity.

The base-exchange capacity of the Gardiners Clay, samples 6 and 12, is clearly higher than that of the Pleistocene sand, in fact approximately in the ratio of 20 to 1. This would suggest that the adsorption of activity in only 10 feet of Gardiners Clay might be as great as that in 200 feet of glacial sand. The experimental and field conditions are sufficiently different, however, so that reliance on such numerical comparisons would be unwarranted.

For example, in most laboratory experiments the clay and the solution are thoroughly mixed. In the ground, however, such intimate contact is not to be expected, for the clay has such a low permeability that little water will flow through it. Further, the water that does pass through is probably largely restricted to certain comparatively permeable zones, so that the adsorptive capacity of the bulk of the formation is not called into play. The differences in mineralogical composition between the two formations strongly substantiates the experimental data, for the Gardiners Clay contains clay minerals of the montmorillonite or vermiculite families that have high exchange capacities, whereas the upper Pleistocene deposits contain the much less effective kaolinite as well as hydrous iron oxide, a poor base-exchange material but an active adsorbent of some radioactive ions.

The opportunity for contact between the ground water and the upper Pleistocene deposits is excellent, however. These deposits are unconsolidated so that the ground water moves through the whole mass of the formation. The rate of movement, which probably does not exceed a foot a day, should provide ample time for adsorption or ion exchange to reach equilibrium. Because of these differences, the relative adsorptive capabilities of the two formations under field conditions are difficult to evaluate.

The unidentified unit at the base of the upper Pleistocene deposits appears to be intermediate in adsorptive capacity between the overlying sand and gravel and the Gardiners Clay. The Cretaceous formations resemble the upper Pleistocene deposits in that the clay is largely kaolinite, but the Cretaceous units contain much more kaolinite. Pound for pound the Cretaceous formations appear to be less effective adsorbents than the Gardiners Clay, but because of their much greater thickness their total effectiveness must be much greater.

SUMMARY OF THE DISCUSSION ON CONTAMINATION OF GROUND WATER

A distinction has been made between two classes of accident involving radioactive fluids, the spill and the leak. In a spill the liquid is all released suddenly and presumably at the surface. Perhaps the most important factor in the event of a spill is the volume of liquid infiltrating per unit surface area. If small, that is less than 1 cubic foot per square foot, considerable time, as much as several years, may be required for the greater part of the activity to reach the water table at a depth of 50 feet. Because of variations in permeability, much more than the average amount may be expected to infiltrate at one or more favored spots, and below these spots it is possible that small quantities of activity may reach the water table promptly.

In contrast, a much smaller filament will descend from a long-continued slow leak, perhaps from a buried pipe or tank, as the contaminant will not spread out over the surface.

The amount of dilution to be expected in the vadose zone following a spill or leak is uncertain, but it is not likely to be large. This would be particularly true of a leak, from which the active liquid would descend through a small cross section of sand. Dilution below the water table is probably not a rapid process and probably has less effect on the long filament produced by a leak. Some dilution would result from an elongation of contaminated liquid in the direction of movement, but such elongation would have little effect on a filament that is already long in that direction. No known hydrologic mechanism exists that would tend to widen such a filament in a homogeneous aquifer. It may become thicker and more dispersed as the result of density differences, but the quantitative operation of this mechanism is speculative at present. The only known process which can result in dilution due to mixing is diffusion along the interface between the contaminated and uncontaminated liquids; and even where accelerated by differences in density or rates of flow in different parts of the system, this process acts slowly and would hardly be effective in reducing the activity in a large mass of water.

As it slowly seeps down and joins the main body of ground water at the water table, a contaminated liquid, if of the same density as water, would probably form at first a long ribbon whose length, in feet, will be related to the number of days since contact was first made at the water table and whose cross section, in square feet, will be approximately equal to six times the rate of leakage, in cubic feet per day.

Various changes in shape may take place as the foreign liquid moves with the ground water. Hydrodynamic processes and adsorption may elongate the shape in the direction of movement. In a homogeneous aquifer, density differences would tend to thicken the body and would possibly make its upper surface more diffuse. On the other hand, in an aquifer made up of horizontal beds of different permeability, the liquid, if denser than the ground water, would tend to spread out on the top of any poorly permeable bed. The counter currents due to the difference in the direction of movement of the sinking liquid and the horizontally moving ground water would create an unstable pattern with at least the possibility of still further mixing.

Adsorption will in all probability greatly delay the movement of the radioactive isotopes. It will delay some isotopes more than others and so effect a separation of the contaminants. Radioactive decay, made more effective by this delay, will greatly lower the level of the radioactivity before the material can move to the boundaries of the Laboratory reservation. Even if not delayed by adsorption, the rate of movement below the water table is so slow that only isotopes with half lives measured in years would be of concern in areas outside the Laboratory tract.

POSSIBLE CONTAMINATION OF SURFACE WATER

The surface of the ground in and near the Laboratory is so permeable that flow over the surface for any distance is impossible. After a heavy rain on soaked or frozen ground, small streams flow for short distances, but it would be impossible for contamination to be carried from the Laboratory into the Carmans or the Forge Rivers in this way.

The sanitary sewer system, which leads to the sewage-disposal plant, is a special problem. The effluent from this plant is discharged into the headwaters of the Peconic River, which has been dug out into a ditch to receive this extra flow. The disposal-plant filters remove the greater part of the minute quantities of radioactivity that occasionally reach them, but their effectiveness in case of a major accident is less certain (Gemmell, 1952). The minute quantities of activity that now find their way into the sanitary sewers, largely from the hospital area, from the bulk of the activity that the Laboratory deliberately discharges under authorized controlled conditions. These amounts could not be significantly increased by an accident at the hospital area, as the total quantities of activity there are never large. The sewers, however, lead to other buildings as well. For example, a branch extends past the waste-storage tanks at the concentrating plant and might be flooded after an accident. Any contaminant reaching the Peconic River will move downstream at a rate which would depend on the stage of the water table and the flow of the river. H. D. Brice and A. W. Harrington of the Geological Survey estimated that the travel from the Laboratory to Riverhead would take a few weeks, but this estimate is only approximate as no measurements were attempted. The rate of travel of surface water, however, is so much more rapid than that of the ground water that lack of time would permit only isotopes with a half life of a few days or less to be reduced significantly in concentration by radioactive decay.

Cationic activity will be removed from a stream by adsorption on suspended particles of clay or on clay in the bed and banks of the stream, and so lower the level of activity. Dilution will take place much more readily in a stream channel than in an aquifer because of turbulent flow in the stream channel. These safety factors, however, are of small importance as compared with those resulting from the much slower rates of travel of radioactive contaminants in ground water.

Much of the ground water under the Laboratory tract moves southwestward or southeastward and appears as effluent seepage in either the Carmans River or the Forge River; a little may reach Moriches Bay. It would take many years for a contaminant to make the journey, but a long half-life isotope that was not adsorbed on the way would probably eventually show up in one of these streams.

In the Laboratory area, each vertical strip of the upper Pleistocene deposits, transverse to the direction of ground-water flow, 1 foot wide and including the full thickness from the water table to the Gardiners Clay (150 ft), carries on the average 0.13 gpm (gallon per minute).

The long ribbon of contamination that would result from a slow leak, if it were 10 feet wide and 50 feet thick, would represent a flow of about one-half gallon per minute. The stream of contaminated water resulting from a major spill, if 100 feet wide and involving the full thickness of the aquifer, would represent a flow of 13 gpm. The average flow of the Forge River at Route 27 is more than 20,000 gpm, so that the dilution factor for the thinner ribbon would be 8×10^3 or 4×10^4 and of the larger stream of contamination 3×10^2 of 2×10^3. Such dilution factors, although inadequate by themselves to alleviate the contamination that might result from a serious leak or spill of highly active fluid, may well serve to reduce the contamination below the danger level when working in conjunction with adsorption and the natural decay of the isotope during its travel. Contamination of a stream is, therefore, less likely than contamination of the ground water, as stream contamination would be a later secondary result of ground-water contamination, except in the Peconic River, where special circumstances prevail.

MONITORING OF WATER RESOURCES

GROUND-WATER MONITORING

What has been said about the probable pattern of movement of contamination has considerable bearing on the problem of sampling the ground water for monitoring. Several different patterns or procedures for this sampling have been used or considered; none is entirely satisfactory, and some are basically unsound.

Monitoring the ground water would serve several purposes. If effectively done it might detect an unsuspected leak or spill. The undetected loss of any considerable volume of liquid or amount of activity is very improbable in view of the many safeguards and precautions that are employed. Also remote is the possibility that an accident would not be reported by the person responsible. Perhaps a more realistic reason for a systematic sampling and analytical program would be its effectiveness in answering any possible accusation that the Laboratory had contaminated the ground water. Early in the work at Brookhaven the writer began to consider how a sampling program might be set up that would make it possible to show even the most skeptical critic that no contamination had taken place.

At Brookhaven there are only a few places where radioactive solutions are used or stored in any quantity. The most important of these is the "farm" of waste-storage tanks, an area about 200 feet square. The original intention was to install one or more 2-inch wells equipped with hand pumps immediately southeast of the farm and to periodically pump small samples of water and test these for activity. The problem was to locate the wells properly so that a minimum number would monitor the maximum area. When it was later realized that a slow leak would probably result in a long, narrow ribbon of contamination, having a width of perhaps only a few feet, the spacing problem showed the plan to be impractical. Twenty wells spaced 10 feet apart would hardly be enough to sample the ground water adequately, but they would be too numerous to use conveniently. The problem of selecting the proper depth for the screens could be met only by the awkward and expensive expedient of screening the full 150 feet of saturated sand down to the Gardiners Clay. In other words, the concept of using closely spaced wells of small diameter to monitor the perimeter of even a small area for a possible leak is impracticable. To monitor even a single tank is difficult, in that over short distances the exact direction of movement of the ground water can be established with certainty, if at all, only by the use of tracers.

The area monitored by a single well, what might be called its effective sampling radius, can be increased by pumping the well, but the possible increase with a small well is not large. Assuming that the surrounding sand has a porosity of one-third, it would be necessary to pump more than a thousand gallons of water from a small well with 150 feet of screen to draw in water from a distance of even 1 foot, and nearly 5,000 gallons to give an effective radius of 2 feet. If the length of screen were smaller, less water would have to be pumped for the same increase in effective radius, but the volumes that would be required to materially improve the effectiveness of any well are far greater than can be pumped regularly by hand.

Early in this study some thought was given to a second approach to the sampling problem. The hope was to pump small samples periodically from each of a line or "picket fence" of wells along the southeastern perimeter of the Laboratory tract. If these wells were spaced closely enough, it was argued, then it would be possible to state categorically that no contamination could possibly have escaped from the Laboratory by way of the ground water. When it was realized, however, that no widening or "scattering" of a narrow ribbon of contaminated water would result from its movement through the aquifer, the close spacing that would be required and the consequent large number of wells showed the plan to be basically unsound and impractical.

A third approach to the sampling problem is to take samples from representative private and public-supply wells in the nearby communities and particularly from wells in the more densely populated neighborhoods. This sort of spot check is of more potential psychological value than scientific, and its possible future usefulness depends on a correct anticipation of the areas from which questions of possible contamination may come. The procedure does appear to have some public-relations value, however, and for this reason the Geological Survey group carried on a sampling program of this general sort at Brookhaven for about 2 years. (See de Laguna, 1964.)

The problem of ground-water sampling is primarily one of properly locating the sampling well, both for position and depth of screen, so that the sample will provide significant information about some point or area of interest. The frequency of sampling is probably not critical. If the ground water is moving at a rate of 1 foot per day, the possibility of missing a body of contaminated water because it passed by between times of sampling is not serious, even if samples are collected at infrequent intervals.

The problem of ground-water sampling at Brookhaven could be solved by a somewhat different approach. The Laboratory pumps water at a rate approximating 500 gpm, using 3 wells in rotation. If all this water came from only well S2476 (Laboratory well 1) and this well were screened from the water table down to the Gardiners Clay, ground water under a fairly extensive area would all be drawn into this one well. The size of this area, which is roughly parabolic, may be estimated approximately as follows: The area will extend upgradient to the ground-water divide, and it will have a width at some distance from the well that just suffices for the aquifer to carry the volume of water the well is pumping. This upper Pleistocene aquifer has a transmissibility of about 200,000 gallons per day per foot and a gradient of 5 feet per mile, so that each foot width of aquifer measured normal to the direction of movement of the ground water carries 0.131 gpm. Therefore, 500 gpm will represent the normal flow through about 3,800 feet width of aquifer. A short distance northwest of the well the artificial gradient due to the pumping will be added to the natural gradient, so that a narrower width of aquifer will carry all the water moving to the well.

Somewhere southeast of the well the natural gradient and the gradient due to pumping will be equal and opposite; this point will mark the southeast limit of water moving into the well. The distance to this point is about 600 feet because a circle of 600-foot radius has a circumference of 3,800 feet, and the gradient due to pumping over a section of this length will be equal to the natural gradient.

A more exact solution is given by the formula:

$$y = -x \cot \frac{2Tix}{Q},$$

where x and y define the limit of water flowing into a well at the origin, with y in the direction of the natural gradient, T is the transmissibility of the formation, i is the natural ground-water gradient, and Q is the rate of pumping. For the limiting value where $x=0$,

$$y=\frac{Q}{2Ti},$$

Q, the rate of pumping, will have to be expressed in gallons per day, if T is given in these units. This formula was supplied to the writer by Herbert J. Kouts of the Laboratory staff, although it was later realized that the same formula had been devised by Theis (1932) in another connection. The map shown in figure 8 was prepared from this formula. The boundary shown includes the area that will contribute water to the well under steady-state conditions after the well has been pumped for an indefinitely long time. The area will be closely approximated, however, after several months of pumping.

The formula assumes that the formation is uniform and hydrologically isotropic and that the movement of the water is two dimensional. This last assumption demands that the well be screened through the full thickness of the aquifer, for the flow lines converging into a shorter screen make the problem far more difficult to solve. If the screen penetrates the whole thickness of the aquifer, it will draw water from the several parts of the aquifer roughly in proportion to their permeability; this is desirable as the ground water in the less permeable beds is moving less rapidly. Thus the influence of the well will be felt at roughly equal distances in both the more permeable and less permeable beds. The possibility of relatively dense contaminated water passing under the screen will also be avoided.

The use of the Laboratory's supply well as a sampling well offers several additional advantages. The procedure would give assurance that could be gained in no other way—that no leak or spill of activity had contaminated the ground water. With only one well to consider, samples could be taken conveniently at regular intervals. Any body of contaminated water would be considerably diluted by passing into the pump, as it would be mixed with the water normally flowing through a cross section of the aquifer 150 feet thick and more than 3,800 feet wide. Most important of all would be the psychological advantage of being able to answer any inquiry about possibly contaminated ground water by showing that the Laboratory was itself using all water subject to possible contamination. The plan, however, would commit the Laboratory to a continuing close supervision of its own water supply.

Even without any alterations, the two Laboratory supply wells, S2476 (Laboratory well 1) and S6697 (new Laboratory well 3) are of some value for ground-water monitoring. The volume of water

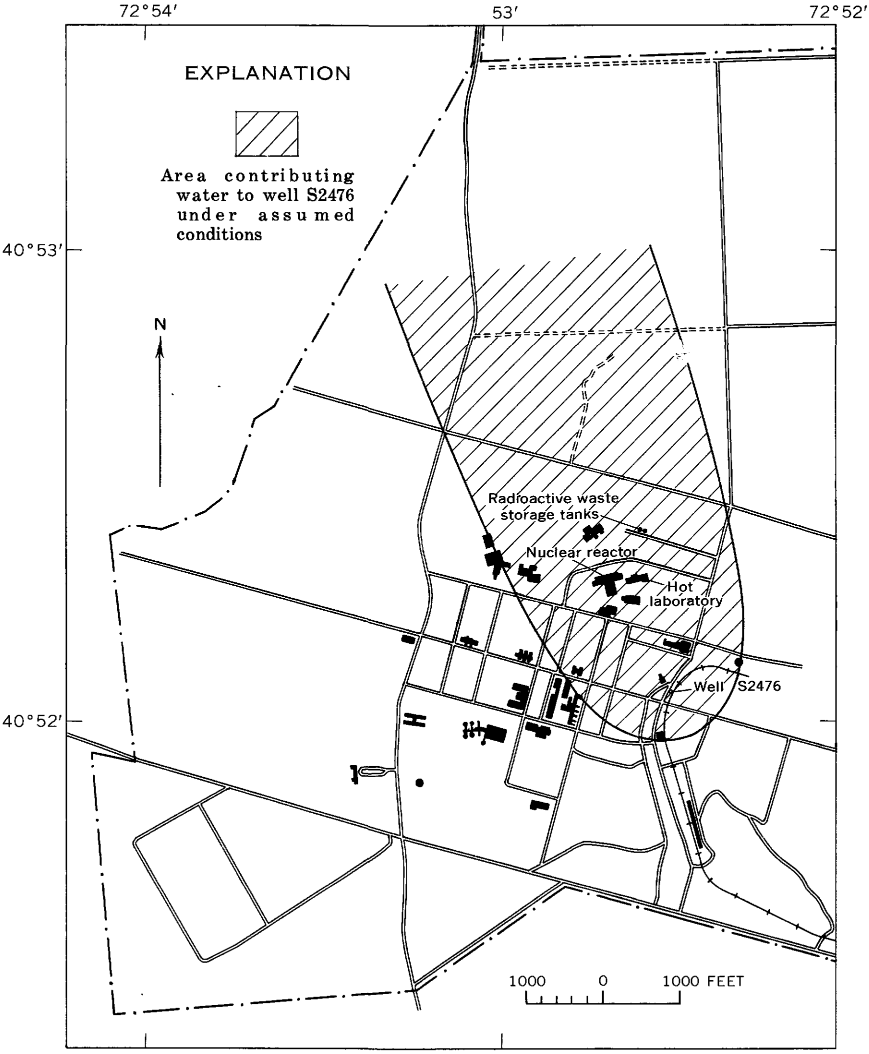

FIGURE 8.—Area contributing water to pumping well S2476 after proposed modifications; assumed rate of pumping 500 gpm.

pumped by these two wells is sufficient so that they would pump in and intercept much or all the ground water moving under the waste-storage-tank area, the pile, and the hot laboratory, if their screens fully penetrated the upper formation of Pleistocene age. As it is, both wells are about 100 feet deep, and the bottom of their screens are roughly 20 feet below sea level. The top of the Gardiners Clay in their vicinity is about 110 feet below sea level, so that approximately 90 feet of saturated upper Pleistocene deposits lies below the bottom of their screens. If the aquifer were not bedded, and if the postulated contaminating liquid were no denser than the ground water, this would make little difference. As it is, the contamination could pass below the wells. On the other hand, some of the diluted contamination would sink less rapidly, so that the sampling of these wells would be desirable, although negative results could not be considered conclusive evidence of no contamination.

SURFACE-WATER MONITORING

The questions of the best procedure for monitoring the several streams near the Laboratory, and the value of such work, depends on other considerations. The greater part of the ground water moving under the Laboratory tract finds its way eventually into either the Carmans or the Forge Rivers. Sampling these streams should show whether the Laboratory was letting any activity escape, for, like the pumping well, the streams receive all the water from a wide area. The streams, however, receive water from such a wide area that the hypothetical ribbon of contamination might well be diluted by several orders of magnitude, and thus require greater analytical sensitivity for detection. A second objection is that by the time the ground water reaches the Carmans or the Forge Rivers it has already passed under a wide populated area. As a warning of ground-water contamination, analysis of river samples would provide information that could be too little and too late.

Contamination entering a stream directly, as at the sewage-disposal plant, would not be sufficiently diluted to make its detection by analysis difficult, if the amount of activity involved were of consequence. Routine sampling of the stream to detect this eventuality involves the question of timing, for a small concentrated dose of activity could move past the sampling point in a comparatively short time.

For these several reasons virtually all ground-water and surface-water monitoring at Brookhaven Laboratory has been discontinued. Occasionally, it will probably be desirable to collect and count samples from the Laboratory's own supply wells, but experience suggests no additional simple procedure that would be of firm scientific value.

REMEDIAL MEASURES

PAVING AREA OF SPILL

The discussion that follows analyzes the possibilities for remedial action in the event of an accidental release of radioactive fluids. The analyses are made without regard to the probability of occurrence of the various types of accidents.

In the event of a spill one of the first jobs would be to prospect between the water table and the surface of the ground in the area of the spill. Presumably, the prospecting would be done with small-diameter holes into which radiation-detection probes could be lowered to determine the spatial distribution of the contaminants; undoubtedly, the determination of the distribution would be complex. The first hazard to be considered would not be the contamination of water supplies, but the problem of airborne radioactive dust originating from the affected surface as it dried out. This problem would not be so important in the event of a leak below ground. In event of an aboveground spill, probably the first step would be to pave or cover the affected surface area as rapidly as possible to hold down the dust. An additional advantage will be derived if this covering is made impermeable to water. Almost certainly, a large proportion of a radioactive fluid lost in any spill would be retained in the vadose zone and could reach the water table only if carried down by later waves of moisture from a surface soaked by rain or snow. Paving the surface and channeling the precipitation away would leave much of this contamination trapped harmlessly in place, although small amounts would continue to drain slowly for a long time. Whether the activity is held in the vadose zone in liquid bound by capillary forces or whether it is adsorbed onto clay minerals or limonite, the advantages to be gained by quickly paving the affected area are so great in relation to the expense that this remedial measure should be undertaken without waiting for a study of details.

MAPPING CONTAMINATION UNDERGROUND

If a serious spill ever occurred, the second step should be a study of the detailed hydrology of the immediate area, as the effectiveness of further remedial measures would depend more on detailed knowledge than on prompt action. Test drilling in the area of the spill would be necessary and would be facilitated if the area had been paved. For the initial drilling, 2- or 3-inch pipe driven with a well point should suffice. The pipe and well point could be driven, without the hazard of bringing any material to the surface, to depths of about 100 feet, sufficient to penetrate past the water table in the Laboratory area. Any one of a variety of existing gamma-ray-counting devices could be used to log these holes, and if the concentration of radioactivity were high enough, semiquantitative estimates could be made of the underground distribution of the radioactive materials. Once the general outline of the contaminated space were determined, the next step would be to take soil and water samples so that the distribution of the several isotopes could be determined; some separation by chromatographic processes is to be anticipated. For this, additional wells, drilled so that soil samples could be collected, would be required.

If careful investigation showed that dangerous quantities of radioactive substances of long half life had reached the water table and were traveling with the ground water, three approaches to the problem appear to be available. They are not mutually exclusive, and all three might be attempted simultaneously. They are scavenging, fixing in place, and the development of alternative water supplies. Fixing in place appears to have at best only questionable value, but the other two procedures are believed to be potentially effective and practicable.

SCAVENGING

Scavenging would involve the recovery of contaminated ground water followed by its treatment or transfer to some less dangerous place. The treatment of the large volumes of liquid that would be involved may or may not be feasible. The removal of contaminated water from the upper Pleistocene deposits and its disposal into the Lloyd Sand Member of the Raritan Formation appears to offer some interesting possibilities; and as an alternative to letting the contamination drift downgradient in the upper Pleistocene deposits, the procedure might have merit.

At present (1955) few wells yield water from the Lloyd Sand Member in Suffolk County, and all these are north of the water-table divide. The nearest supply well screened in the Lloyd Sand Member is at Kings Park, about 20 miles west-northwest of the Laboratory. Test wells at Port Jefferson and Brookhaven Laboratory penetrated Lloyd deposits of apparently low permeability. At Greenport, on Orient Point, the Lloyd Sand Member is apparently only 20 feet thick, and at Orient State Park it was found to contain salty water. Thus the Lloyd Sand Member is probably unsuited for water supply over much of Suffolk County, although it is extensively developed in Nassau and Queens Counties to the west. However, present knowledge is inadequate to prove this conclusion.

Very little is known of the natural movement of water in the Lloyd. The recharge area, beyond doubt, is along and adjacent to the water-table divide, but the location of the discharge area is less well understood. The hydrostatic pressure in the Lloyd is hardly sufficient to force fresh water more than 2,000 feet below sea level unless the salty water in the deep deposits off the south shore of the island is less salty and hence less dense than sea water. In any event, fresh water circulating in the Lloyd must begin to move upward somewhere between the Laboratory and the ocean beach.

The water in the Lloyd in the Laboratory area moves in a more easterly direction than that in the upper Pleistocene deposits. Accordingly, if contaminated water were injected into the Lloyd at the Laboratory, it would move in the general direction of Eastport or Westhampton. Where such water might emerge from the Lloyd is not known, nor can predictions suggest whether it would enter overlying water-bearing deposits that were being used or enter salty water beyond the fresh-water–salt-water interface. However, such contaminants would remain in the Lloyd and the overlying poorly permeable deposits for a long time and might be fixed at depth by adsorption.

Assuming that the hydrology of the Lloyd Sand Member were well enough known so that the consequences of disposal of contaminated water into the Lloyd could be evaluated, suitable injection wells could probably be developed in the aquifer to make the plan practicable, even though the permeability of the Lloyd in the Laboratory area is low. For example, tests on well S6434, the larger diameter deep test well at the Laboratory, showed that approximately 250 gpm could be pumped into the Lloyd. The exact amount would depend on the length of screen and the development of the particular well and on the head developed by the injection pumping. With time, the possible rate of injection would decline as the well screen plugged and as the head in the Lloyd in the immediate vicinity of the well increased. Recharge wells used to return water pumped for air conditioning are commonly cleaned when they get plugged, but it might be cheaper to drill a new well rather than to attempt to recondition one into which large quantities of radioactive fluids had been discharged. Although expensive, the cost of such an operation, if successful, would be insignificant in the event of a major accident requiring remedial measures of considerable magnitude. Indeed, following such an accident, cost would not preclude the use of several recharge wells either simultaneously or in sequence, so as to distribute the load and to provide opportunity for repair or replacement.

The assumed figure of 250 gpm for one well is conservative and may properly form a basis for calculations. Consider what might be accomplished by the removal of contaminated water from the upper Pleistocene deposits at this general rate. The pumping well could be placed some distance downgradient from the scene of the postulated accident and screened through the entire thickness of the glacial sand; this is the same scheme that was suggested for the ground-water-sampling well. Pumping at a rate of 250 gpm would slowly form a depression in the water table that would eventually bring into the well all the ground water in the upper Pleistocene in a large area, with a maximum theoretical width of nearly 2,000 feet—just half the size of the area shown in figure 8. Because of variations in permeability in the aquifer the actual size of the contributing area would be somewhat smaller, but still more than enough to include all the area that would be involved in any credible accident. The disadvantage of this arrangement is that the pumping would have to be continued for a very long time because the rate of movement of the ground water in the contaminated area would be little increased over its natural rate of half a foot per day.

A second procedure would be to disregard the natural movement of the ground water and drill the scavenging well in the middle of the postulated contaminated area, screening only that port of the well that was in contaminated water. In the immediate vicinity of the pumping well, the ground water would move radially toward the well. At 250 gpm, the natural ground-water movement would be reversed up to a distance of 300 feet in the direction of the natural ground-water flow, and by screening only a part of the thickness of the aquifer, the scavenging could be made to reach out somewhat farther in this direction at the depth of the screen. An area 600 feet in diameter might well include all the area involved in even a large spill. A cylinder of saturated sand 600 feet in diameter and 150 feet thick contains roughly 100 million gallons of water, and if pumped at a rate of 250 gallons per minute, at least 280 days would be required to reclaim it. In practice, pumping for a year or more would be required to scavenge all the water from such a volume, and such factors as adsorption and inhomogeneity of the aquifer might increase the time appreciably. Even at moderate rates of pumping, however, areas of the order of magnitude that are being considered could be swept clear in a period of time that is long but probably not impractical.

Putting the scavenging well squarely in the middle of a contaminated area involves drilling in contaminated ground. This operation, unlike driving small-diameter observation wells, would require bringing to the surface considerable quantities of the contaminated sand, gravel, and water while drilling and developing the well. Dirty or silty water pumped down the disposal well would quickly clog the well; therefore, the scavenging well would have to be thoroughly developed. The drilling and developing of a well in contaminated material and the disposal of the drill cuttings and silty water would be expensive and laborious but not impossible.

Probably the best procedure would be to drill several wells a short distance downgradient from a contaminated area. Carefully controlled pumping from the wells could then pull in contaminated water as it came by, and could also be used to steepen the water-table gradient locally and so greatly speed up the natural rate of movement. Small-diameter observation wells could be used to determine, by water-level measurements, the exact area contributing water to the scavenging wells, so that the operation could be carefully guided. The pumping of some uncontaminated water along with the contaminated would not be altogether undesirable, for this would provide dilution, and the overall procedure appears practical.

FIXING IN PLACE

The possibility of fixing in place the radioactivity lost in a major accident has come up so often in informal discussion that it appears desirable to advance here the arguments that suggest that the method will not work. Obviously, either the water containing the radioactive substances or the activity itself might be fixed. Fixing the water below the water table would require building a cofferdam around it. To attempt to drive steel piling to a depth of 200 feet does not appear to be possible; to enclose an area of a hundred feet, or of several hundred feet, in diameter in this way would always be very expensive. To sink a barrier of interlocking caissons would also cost a large amount. Another possible approach would be to build up an underground dam by injecting from the surface through pipes or wells a material that would plug the pores of the sand and make the sand impermeable. One proposed method of this sort uses sodium silicate and calcium chloride; others bentonite or cement. One promising process uses an asphalt emulsion. Early trials with this material were encouraging, but later work, done in cooperation with the New England Division, U.S. Army Corps of Engineers, showed that the emulsion was difficult to inject uniformly, as it tended to spread out only in the more permeable beds (J. O. Izatt, the Shell Oil Co., written commun. 1953). This, or a related process, may have value for other special purposes, but it does not appear to be possible to build up an extensive underground barrier to the movement of ground water in the Brookhaven area by any known practicable method.

A second approach would be to fix the activity but not the water. This might be done by injecting into the formation an adsorbent or reagent with a specific affinity for the more dangerous isotopes. Apart from the question as to whether such a reagent exists is the question of getting it into place so that it would mix with or intercept the contaminated water. Here again the tendency of an injected solution to seek out and follow one or more specific beds, to the exclusion of the neighboring beds, would make the uniform emplacement of the reagent almost impossible. A solution of the reagent, deliberately concocted so that it would have a high specific gravity and injected just above the water table or poured out on the surface just downgradient from the spill, would soak down and sink through the ground water, but no thorough mixing would take place.

A final argument may be that the volumes and quantities of material involved make the whole general plan impractical. A volume 100 feet square and 150 feet thick contains 1,500,000 cubic feet. The pore spaces in such a volume of sand would have a volume of 500,000 cubic feet, or 3,750,000 gallons. This is a large volume to surround with a dam or to treat with a reagent.

ALTERNATIVE WATER SUPPLIES

If the ground water in the upper Pleistocene deposits under the Laboratory tract should become generally contaminated with radioactive material, it would be perhaps necessary and probably advisable to get the Laboratory's supply from some other source. Although it is possible to calculate the area from which a well is deriving its water and feasible to check these calculations with measurements in the field, some element of uncertainty would remain owing to the possibility of gravel lenses of much more than average permeability. Through such hidden lenses, some water might be pulled in from a greater distance than had been anticipated. For this reason, relocation of the laboratory's supply wells to some part of the upper Pleistocene aquifer that could remain safely unaffected may require the selection of a site at a considerable distance.

An excellent alternative source would be the basal coarse-grained beds of the Magothy(?) Formation penetrated at depths of about 1,000 feet in both deep test wells at the Laboratory. A well capable of yielding 500 to 1,000 gpm from this material would be in no way unusual, as many public-supply wells in Nassau County pump at this rate from greater depths. The water in the lower part of the Magothy(?) below the center of the Laboratory probably infiltrated into the ground northwest of the Laboratory tract and presumably, therefore, well outside any area susceptible to contamination. A well screened in these deposits below the center of the Laboratory tract and yielding 500 to 1,000 gpm would eventually draw in water laterally from a considerable area, probably several times as large as the area that would contribute to a well of similar capacity screened in the upper Pleistocene deposits. Even after a long period of time, however, it is unlikely that any contaminated water could be pulled in from above. The Gardiners Clay and the clay beds within the Magothy (?) would greatly inhibit any movement directly downward to the lower part of the Magothy (?) from the upper Pleistocene, unless the total water pumped from the Magothy (?) greatly exceeded any present or probable demand. Communities such as Center Moriches or Mastic could, if necessary, be supplied in the same way from deep wells. For a private domestic-supply well, however, which is pumped only for short periods at a rate of 10 to 15 gpm, the upper beds of the Magothy (?) may well be a safe source, as the Gardiners Clay would almost certainly provide sufficient protection.

GENERAL CONCLUSION

In the unlikely event of the loss of a substantial quantity of radioactive solution, the points of major interest will be the direction and rate of movement, the dilution of the radioactive solution with ground water, and the adsorption of the activity by soil materials; these are the factors that will determine the time of arrival and concentration of the contaminant and, consequently, the degree of hazard. Much more is known about the possible operation of some of these factors than others.

At the Brookhaven Laboratory, it is possible to predict the direction of movement of the ground water to within a few degrees and the rate of movement, over all but short distances, within a factor of perhaps two or three. In many geological environments the direction of movement of the ground water may be estimated without serious danger of material error, but the rate of movement cannot be predicted because it is so different in different places. For example, a generally solid and nearly impervious limestone may be traversed locally by solution channels through which ground water may be moving at a velocity comparable to that of surface streams. In any rock in which the permeability is due to fractures, the accidents of geometry or the concentration of stress along certain fracture patterns may produce paths through which ground water may move at rates out of all proportion to its average velocity. The possibility of predicting the average rate of movement of the ground water at Brookhaven at the site of a hypothetical spill is a consequence of the homogeneity of the aquifer that underlies the Laboratory; and because of this homogeneity the rate of movement, even locally, is unlikely to differ significantly from the average rate.

The direction and rate of movement give the time that will be required for the ground water to reach a point of danger and, therefore, the absolute minimum time that will be available for radioactive decay to reduce the concentration of unadsorbed activity. The rate of such decay is, of course, invariably for any isotope; and consequently as the number of half-life periods of the decaying radioactive substances in transit increases, the concentration of activity is reduced by a proportionate geometrical progression. At Brookhaven Laboratory, the transit time to the nearest sensitive points outside the Laboratory tract is measured at least in years and probably in decades, so that only a comparatively few of the longest lived isotopes are a potential hazard.

Dilution, the second major factor, would probably materially reduce the concentration of radioactivity following a small spill but would become proportionately less effective as the size of the spill would increase. As compared to radioactive decay, the difference is between a geometric and an arithmetic progression. The laminar flow of ground water precludes mixing by turbulence; and although density differences and even minor inhomogeneities in the aquifer will result in some mixing, these processes do not appear capable of providing important safety factors in the event of large-scale ground-water contamination.

The affects of adsorption are extremely difficult to evaluate. They depend in part on the rate of movement of the solution, but principally on the clay minerals present, the surface area exposed to the solution, and the concentrations of the various nonradioactive ions in solution.

The environment at Brookhaven is in certain respects, particularly favorable for the reduction of contamination in the ground water by adsorption. Pound for pound the sands that underlie the Laboratory are not particularly active adsorbents, as compared, for example, with shale, but it is not the effectiveness of the aquifer on a pound-for-pound basis that is the most important consideration. Even a few hundred feet of a poorly adsorbent aquifer, such as a glacial sand, has more than adequate potential adsorptive or ion-exchange capacity to take up the contaminants released by even a very large spill. The question is whether the full capacity of the aquifer can be brought to bear or whether the contaminated liquid will move largely through a few selected channels and avoid the bulk of the material.

A highly porous and generally permeable aquifer like the upper Pleistocene deposits at Brookhaven meets the safety requirement fully as the result of those qualities that ensured a slow and uniform rate of movement of the ground water. Our general conclusion, therefore, may be that Brookhaven National Laboratory is in a geologic and hydrologic environment that is unusually well suited to reducing the dangers of serious contamination of the ground water in the event of an accident releasing large quantities of radioactive solution.

RECOMMENDATIONS FOR FURTHER STUDY

Probably the most valuable addition to the present knowledge of water contamination would be information on the capabilities of the formations to adsorb radioactive isotopes. Only certain of the isotopes require detailed study, for only those wtih a half life measured in months or years would represent a hazard at Brookhaven. Even a little information would indicate the relative importance of sorption as a factor in retarding the movement of dangerous contaminants.

Direct measurements of the rate of movement of the ground water in the Brookhaven area would be desirable to supplement the conclusions from an analysis of pumping-test data. Direct measurement of velocity by tracers would furnish confirming data and might also provide some idea of the variations in velocity with depth and location.

Of somewhat less direct concern to the subject of this report would be a study of the hydrodynamic processes that may provide some measure of dilution. More specifically, it would be of interest to know the degree of dilution to be expected as a function of the hydraulic properties of the aquifer and of the distance traveled. Better experiments of the type shown in figure 1 should provide at least a partial answer.

The initial phase of any accidental liquid discharge would involve movement through the zone of aeration. At present even the best methods for the measurement of soil moisture are inadequate to give a clear picture of the annual cycle of ground-water recharge. Better quantitative understanding of this natural cycle and of the movement of liquid that may be lost in a spill will require not only better instruments but also a better practical quantitative theory of the movement of water through unsaturated media.

REFERENCES

de Laguna, Wallace, 1964, Chemical quality of water, Brookhaven National Laboratory and vicinity, Suffolk County, New York: U.S. Geol. Survey Bull. 1156–D, 72 p.

Faust, G. T., 1963, Physical properties and mineralogy of selected samples of the sediments from the vicinity of the Brookhaven National Laboratory, Long Island, New York: U.S. Geol. Survey Bull. 1156–B, 34 p.

Gemmell, Lee, 1952, Efficiency of filter beds for treating radioactive waste: Nucleonics, v. 10, no. 10, p. 40–42.

Lapidus, Leon, and Amundson, N. R., 1952, The effect of longitudinal diffusion in ion exchange and chromatographic columns, [pt.] 6 *of* Mathematics of adsorption in beds: Jour. Phys. Chemistry, v. 56, p. 984–988.

National Bureau of Standards, 1953, Maximum permissible amounts of radioisotopes in the human body and maximum permissible concentrations in air and water: Handb. 52, 45 p.

Theis, C. V., 1932, Equation for lines of flow in vicinity of discharging artesian well: Am. Geophys. Union Trans. 13th Ann. Mtg., p. 317–320.

Watkins, J. Wade, 1954, Use of radioactive iodine as a tracer in water-flooding operations: Am. Inst. of Mining and Metall. Engineers Tech. Pub. 3894.

Wind, R., 1951, Een methode von het verkrijgen van uit zandgronden, *in* Water: The Hague, Netherlands, v. 35, no. 4, p. 35–37; abstract by Rudolfs, W., August 1951: Jour. Am. Water Works Assoc., v. 43, no. 8, p. 52.

U.S. GOVERNMENT PRINTING OFFICE: 1966 O—799-427

[redacted]