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MOVEMENT OF RADIOACTIVE EFFLUENTS IN NATURAL WATERS AT HANFORD

By

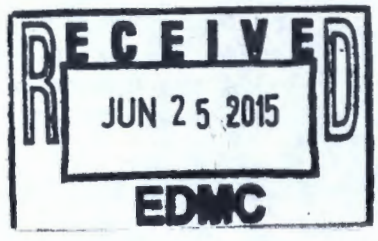
J. F. Honstead, R. F. Foster, and W. H. Bierschenk

Conference on the Disposal of Radioactive Wastes

Principality of Monaco

November 16-21, 1959

Hanford Laboratories
Hanford Atomic Products Operation
General Electric Company
Richland, Washington



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INTRODUCTION

The hazardous quality of radioactive isotopes cannot be destroyed except by means of natural decay. The processes commonly referred to as waste "disposal" therefore, actually resolve into systems which perform one of two functions. Either the radioactive material is injected into a path that is of long enough duration to permit suitable decay of the radioisotopes before they appear in a populated domain, or they are sufficiently diluted by environmental materials to levels of no concern to the population. In actual practice, a combination of the dilution-decay principles is generally employed.

At Hanford, there are two major sources of low-level radioactive effluent solutions. One of these is the large volume cooling water stream discharged from the reactors. The radioactive material in this water results from the activation of impurities in the water and corrosion products from the reactor hardware. The activation products are characterized by relatively short half-lives and are generally less hazardous than the long-lived fission products. The cooling water is discharged directly to the Columbia River where the very large dilution capacity and a significant decay interval reduce the concentration of radioisotopes to a small fraction of the maximum permissible drinking water limit for members of the public living in the neighborhood of controlled areas.

The second major source of low-level activity solutions is the complex system of chemical plants used for the processing of uranium fuels to recover the plutonium product. The radioactive material contained in these wastes is largely a mixture of fission products. Typically, the relative hazard of a mixture of fission products is determined to a major extent by its concentration of strontium-90 (28 year half-life). Since

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this isotope is very toxic, the wastes must either be decontaminated with respect to Sr^{90} with exceptional efficiency or must be disposed of in a manner that assures a suitably long decay interval before they reach a point accessible to the public. Controlled ground disposal of low activity solutions from Hanford chemical processing plants utilizes primarily the long decay interval concept of protecting employees and neighboring populations from exposure to radioactive materials. Thus, wastes are permitted to infiltrate the earth sediments and those components that eventually reach the water table move slowly with the ground water to the Columbia River.

The Hanford plant is located in southeastern Washington in a semi-arid region within the rain shadow of the Cascade Mountains. The more than 1,500 square kilometers controlled by the AEC at this location provide ample isolation from populated areas (Figure 1). The Columbia River flows through the plant providing large supplies of water for plant operation and dilution for low-level waste effluents. The Columbia flow measuring station nearest the Hanford reactors is some 98 kilometers upstream. The runoff from 232,000 square kilometers flows by this station (33). There is normally one major temperature cycle and one major flow cycle annually on the Columbia River. The river generally crests in the month of June with flows of more than 15,000 cubic meters/sec. and it has a low water period which extends from September to April with minimum flows of about 1700 cubic meters/sec. River temperatures are typically highest, about 20° C, in August and September as the high flow recedes. Low river temperatures, about 2° C, occur rather regularly in February. Inasmuch as over much of its course the river bottom is sandy or rocky, the water is clean with turbidities generally averaging less than 7 ppm except during high flows when they may briefly exceed 30 ppm. The hydrology of the area is such that all underground drainage beneath the plant is directed toward this river (1). The river thus represents the sole path whereby radioactive material in solution can be transported into the public domain. After leaving the plant, the first locations at which the waters of the Columbia River are used for human consumption in large quantities are the cities of Kennewick and Pasco. A few minor applications of river water upstream of these cities for irrigation, fishing, bathing, and boating are also subjected to radiological study.

EFFLUENT DISPOSAL TO THE RIVER

Of the radioactive wastes generated by the operation of the Hanford reactors, the predominant ones are reactor cooling water streams. After passing through the reactor and picking up heat and radioactive contamination, the spent coolant -- reactor effluent -- is discharged into the river following a one-to-three-hour holdup period in retention basins.

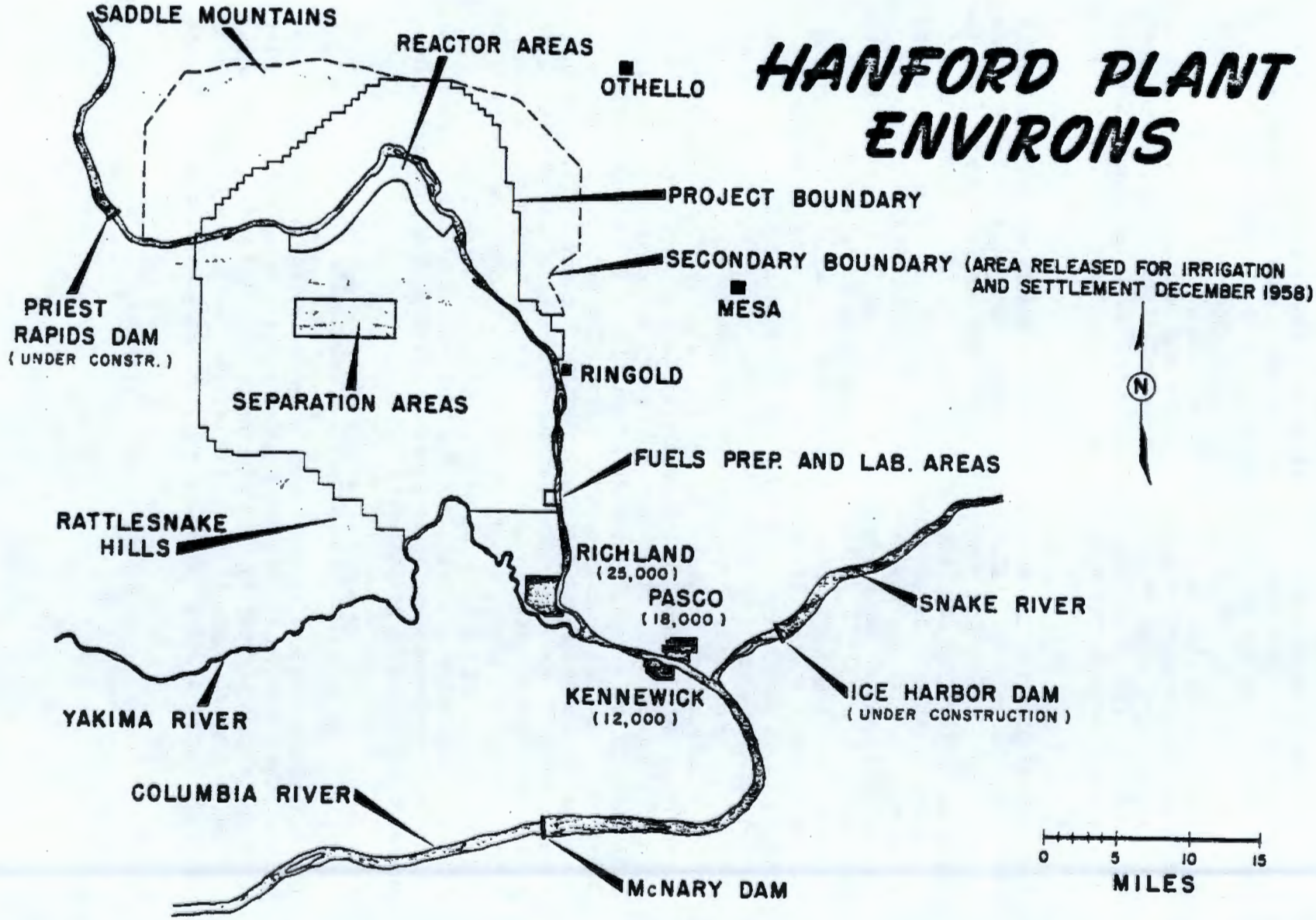

HANFORD PLANT ENVIRONS
SADDLE MOUNTAINS
REACTOR AREAS
OTHELLO
PROJECT BOUNDARY
SECONDARY BOUNDARY (AREA RELEASED FOR IRRIGATION AND SETTLEMENT DECEMBER 1958)
MESA
PRIEST RAPIDS DAM (UNDER CONSTR.)
SEPARATION AREAS
RINGOLD
FUELS PREP. AND LAB. AREAS
RATTLESNAKE HILLS
RICHLAND (25,000)
PASCO (18,000)
SNAKE RIVER
KENNEWICK (12,000)
ICE HARBOR DAM (UNDER CONSTRUCTION)
YAKIMA RIVER
COLUMBIA RIVER
McNARY DAM
N
0 5 10 15
MILES

Figure 1

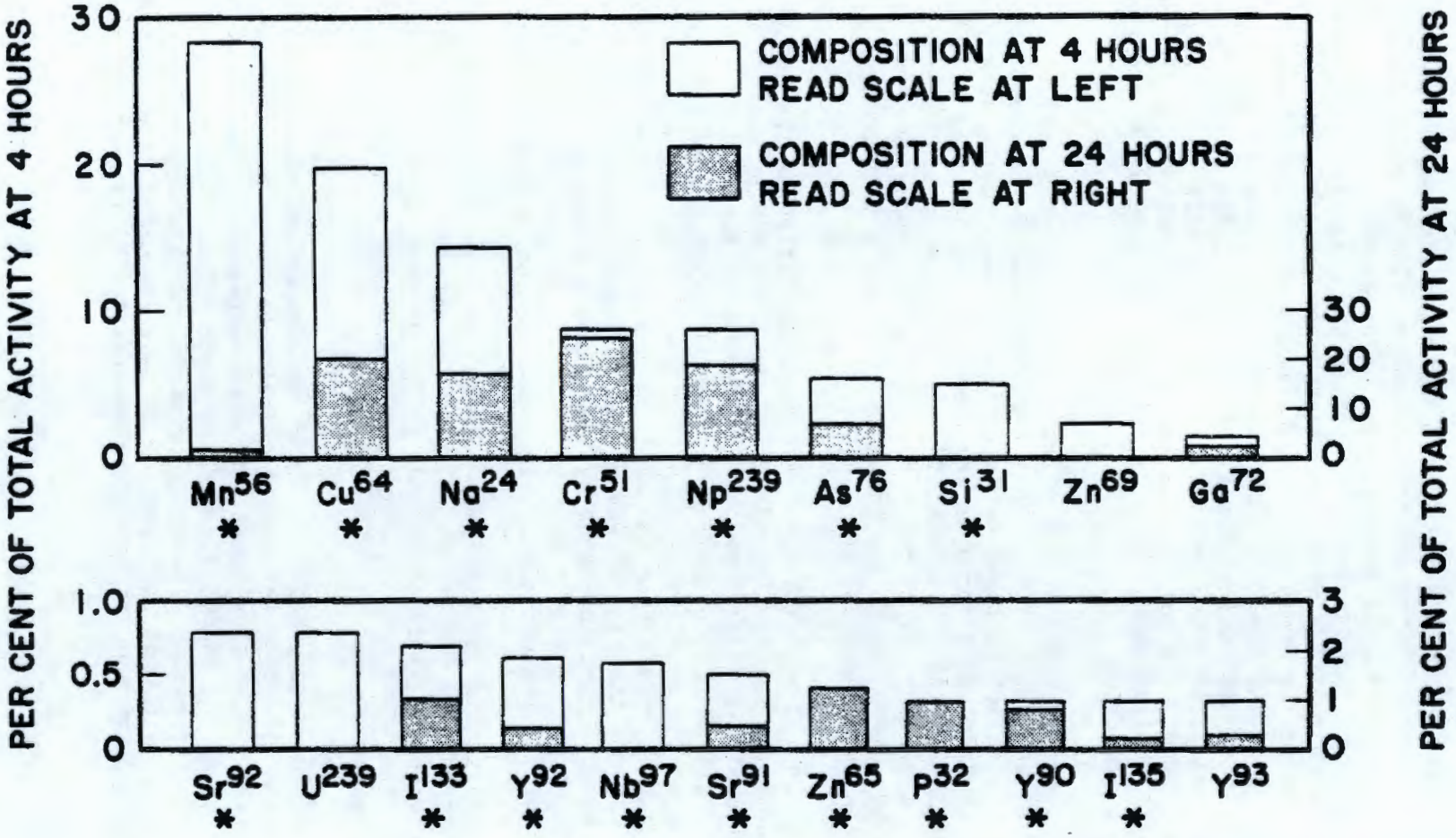
Radioactive isotopes are formed in the reactor by the neutron bombardment of material present in the neutron flux by mechanisms that may involve several nuclear reactions. For example, sodium is a common constituent of the cooling water fed to the reactor, and a substantial part of the radioactive sodium-24 measured in the effluent originates from this source by means of an (n, γ) reaction. Sodium-24 can also be formed from an (n,α) reaction with aluminum, and since aluminum is used for jackets of the fuel elements and for some of the reactor piping, a significant part of the radiosodium comes from this source. Another source that contributes part of the sodium-24 is a magnesium isotope impurity in the cooling water which is converted to sodium-24 by means of an (n, p) reaction.

Although the exact source of many of the important effluent isotopes is as yet uncertain, it is known that the bulk of the radioactive materials are activation products of impurities present in the water, of elements in the film which forms on the reactor tubes and fuel elements, or of constituents in the metals of which the reactor components are constructed. Some minor amounts of fission products also occur as a result of natural uranium dissolved in the Columbia River water undergoing a fission reaction in the active zone of the reactors.

More than 60 radioisotopes have been identified in the effluent. Figure 2 shows the relative abundance of those comprising most of the total radioactivity under normal circumstances at four hours and 24 hours after irradiation. Since the rate of radioactive decay is different for each isotope, the relative abundance of the various isotopes is constantly changing. By the time the effluent has traveled some 55 kilometers to the vicinity of the city of Pasco, the effluent composition might be expected to resemble that indicated by the shaded portions of the bars in Figure 2. Of the total number of isotopes detected in the effluent, 24 are of sufficient interest that their concentration is measured on a routine basis.

The travel time of the river between the reactor effluent outfall furthest downstream and the intake of the Pasco city water plant (about 55 km) has been measured by float methods (30). Minimum travel times were 22.4 hours at a river flow of 2500 cubic meters/sec. and 11.2 hours at 10,000 cubic meters/sec.

The effluent discharged to the river becomes uniformly distributed in the vertical direction relatively quickly because of the force with which it leaves the discharge pipes, its high temperature, the turbulence of the river, and the relatively shallow depth - 7 to 14 meters The effluent is slowly spread horizontally by turbulent diffusion through the 300 to 600 meter width of the river as it is transported downstream (22). A typical dispersion pattern from one reactor is illustrated in Figure 3.



RADIOELEMENT CONTENT OF REACTOR EFFLUENT 4 HOURS AND 24 HOURS AFTER IRRADIATION. (Radioelements marked * are routinely measured).

Figure 2.

TYPICAL DISPERSION PATTERN FOR EFFLUENT FROM ONE REACTOR

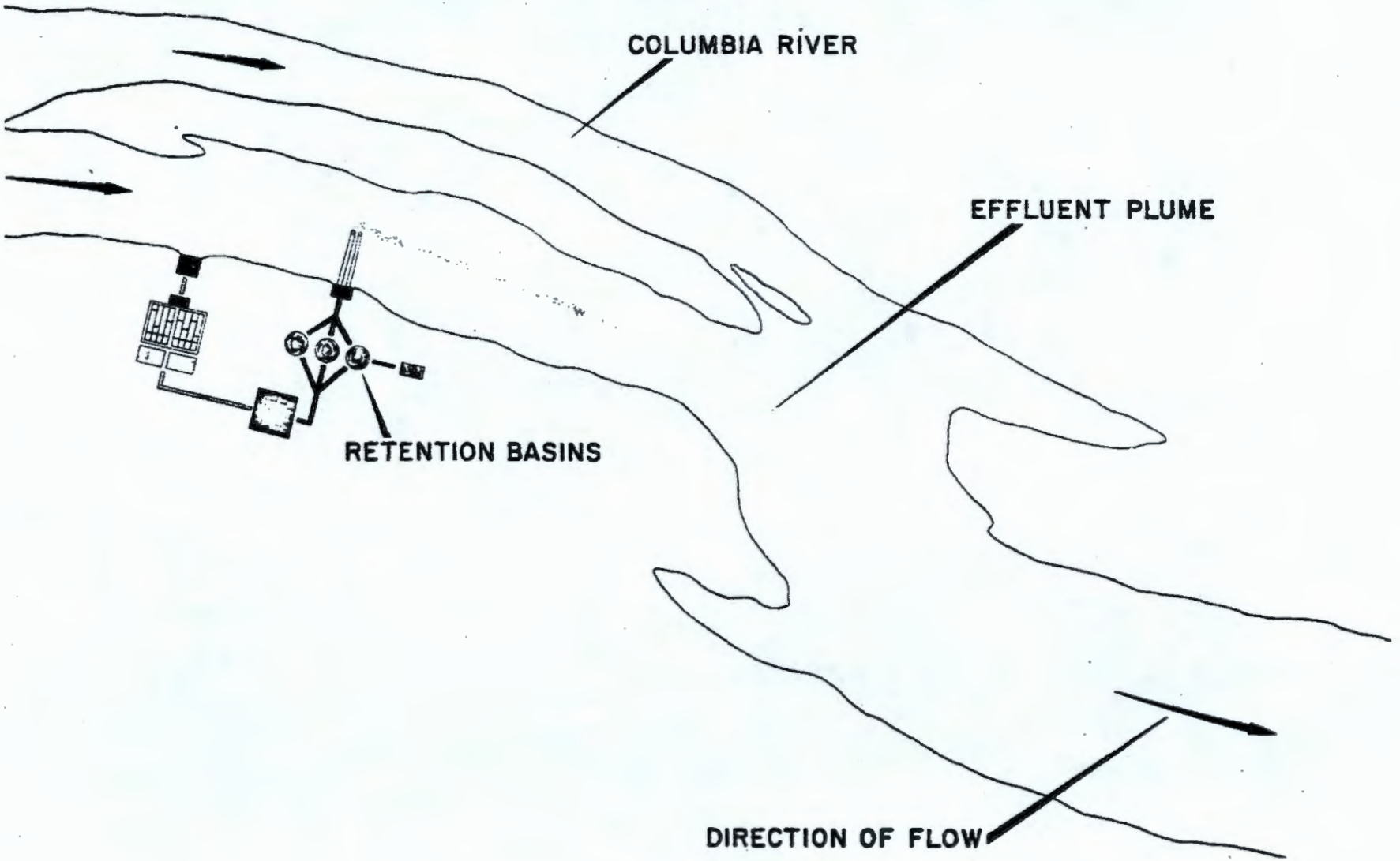


Figure 3

The tendency for high concentrations of the effluent to remain in midstream is advantageous to downriver reactors since their shoreline intakes pick up a smaller fraction of the radioactive material. As mentioned previously, during the fall and winter months the river flow is at its lowest, while during the spring freshet the flow may increase several times and thus reduce the travel time between the reactors and the city of Pasco. While this allows less time for radioactive decay of the effluent isotopes, the potential exposure from drinking river water is actually reduced because of the greater volume of water available for dilution.

The measured amount of radioactive materials in the river water at Pasco is less than that calculated from the quantities released from the reactors' retention basins and the measured travel times. This is ascribed to the scavenging of certain isotopes by living organisms, suspended silt particles, and bottom sediments. This depletion may be considered as a type of "self-purification" which has significance in the estimation of the radiation exposure to people who drink the water and in accounting for the accumulation of radioisotopes in aquatic life and river sediments. After correction for radioactive decay, the average depletion amounts to about 40% between the effluent discharge points and Pasco. The degree of depletion is different for each particular isotope, values having been measured for the 16 individual isotopes which make up 98% of the effluent activity. Depletion varies from less than 10 to more than 60%. Since this depletion is largely dependent upon the solids content of the river, it is to be expected that the self-purification of the river will vary with stream conditions. Losses of individual isotopes from the water indicate that chromium-51 and zinc-65 should be prominent in river sediments and this has been confirmed. No accumulation of long-lived fission products has ever been found in the Columbia River.

At the present time radioisotopic determination of about 20 isotopes or groups of isotopes (sufficient to define the exposure accurately) are made at weekly intervals on samples of water collected from the Columbia River immediately below the reactors and at Pasco, and from the drinking water supplies of Pasco, Kennewick, and the most downriver reactor. These weekly analyses are augmented by analyses of samples obtained from automatic equipment which collects and combines aliquots over a week's period. The composite samples are especially useful in determining the average concentration of the longer-lived isotopes. The contribution which each reactor makes to the total isotope load in the river is determined from samples of the effluent which are analyzed each day for gross beta and alpha emitters, each week for uranium, plutonium, and polonium, and twice each month for the full 20 isotopes or groups which are of interest to the human exposure limit.

From the comprehensive biological surveys of the river carried out during the early years (12)(26), it was possible to select a few key organisms such as the Rocky Mountain whitefish, and representative sampling sites

to permit continued routine assay of the concentration of effluent isotopes in the aquatic forms. Such measurements are now carried out as a part of the regular monitoring program which is used to evaluate the radiation exposure associated with the operation of the Hanford plants (20).

LIQUID WASTE DISPOSAL TO THE GROUND

Operation of the Hanford chemical processing plants requires 1.5 to 2.3 x 10^{10} liters of water per year, all of it being supplied from the Columbia River. A small fraction of this water leaves the chemical processing plants in high-level radioactive waste streams to be stored in underground tanks, but most of it leaves as low and intermediate-level radioactive wastes to be disposed of directly to the ground. Some 70 meters of sand and gravel of glacial origin (glaciofluviatile sediments) immediately underlie the waste disposal sites (6). Below this is an earlier fluviatile or lacustrine formation (Ringold formation) which is as much as 200 meters thick, consisting of silts, sands, and gravels with several clay beds. The water table which is from 70 to 120 meters below disposal sites lies largely within the Ringold formation but extends in some places into the overlying glaciofluviatile sediments. Below these two major units is the relatively impermeable basalt.

According to Hanford definition, the radioactive concentration of intermediate-level wastes ranges from about 10^{-5} to 100 μc/cc. Chemical processing plant wastes in this category have included secondary decontamination streams, condensates from high-level waste concentrators, and supernatant effluent from scavenging processes in aged high-level waste tanks. Over 10^{10} liters of such wastes had been admitted to the ground through mid-1959. Separations process cooling water and utility steam condensates which may occasionally be slightly contaminated from process vessel leaks are considered in the category of low-level wastes with radioactive concentrations less than about 5 x 10^{-5} μc/cc. About 1.4 x 10^{11} liters of such water have been discharged to ground.

At Hanford, the semi-arid climate (averaging less than 18 cm of rainfall per year), a suitable fraction (1-5%) of montmorillonitic clay distributed through the permeable surficial sediments, and the deep water table combine to produce a situation wherein most of the long-lived radioisotopes in the waste are trapped by electrochemical bonds or are immobilized by other reactions as the liquids seep downward through the soils. Those wastes that reach the water table move with the ground water toward the Columbia River, the direction and rate of movement being dependent upon the hydraulic characteristics of the transmitting aquifers.

The hydraulic characteristics of Hanford aquifers have been measured and estimated by a variety of established field methods (2). These include evaluation of data from pumping tests, specific capacity tests, tracer

tests, cyclic ground-water response to river level fluctuations, and hydraulic gradients. Mutually consistent results show that the permeability of the glaciofluviatile sediments ranges from about 0.46 cc/sec/cm^2 (unit gradient) to more than 2.8 cc/sec/cm^2 and the permeability of the underlying Ringold deposits ranges from about 4.6×10^{-3} to 2.8×10^{-2} cc/sec/cm^2 (2). A summary of results is given in the following table.

AVERAGE FIELD PERMEABILITY, cc/sec/cm^2 (unit gradient)

Aquifer tested	Pumping tests	Specific capacity tests	Tracer tests	Cyclic fluctuations	Gradient method
A	0.60 - 3.1	0.46 - 3.0	2.9	0.79 - 2.7	---
B	0.042 - 0.23	0.046 - 0.19	---	0.046 - 0.28	---
C	0.0023 - 0.028	0.0028 - 0.014	---	0.007 - 0.023	0.0046 - 0.014

A - Glaciofluviatile, B - Glacial and Ringold, C - Ringold

Since 1944, the chemical processing plants have discharged to ground over 1.4×10^{11} liters of liquid effluents. Such large volumes have had a profound effect upon the regional water table. Figure 4 shows the contours on the undisturbed water table interpreted from the earliest measurements of water level in wells and from general hydrological knowledge. Similar maps have been prepared periodically over the years and with increasing detail as more wells became available (4). The ground-water contours as of June 1959 are shown in Figure 5. Two distinct ground-water mounds have been created on the water table, their location, elevation, and shape determined by the location of the disposal sites which feed them and upon the nature and geological attitude of the sedimentary formations in which they occur. It is of importance to study these mounds since they determine the direction and rate of flow of the ground water, and this in turn is important in the proper location of disposal sites and in following the underground movement of mobile materials.

In the absence of more precise data, it is assumed that ground water always moves in the direction of the hydraulic gradient indicated by the water table contour map. Therefore, the best means of determining the direction of movement is by drawing vectors perpendicular to ground-water contours from high to low head. Strictly, even a perfect contour map of the water table would show only the horizontal direction of movement of the ground water at the water table. The hydraulic gradients are three dimensional, however, and the water moves not only along the

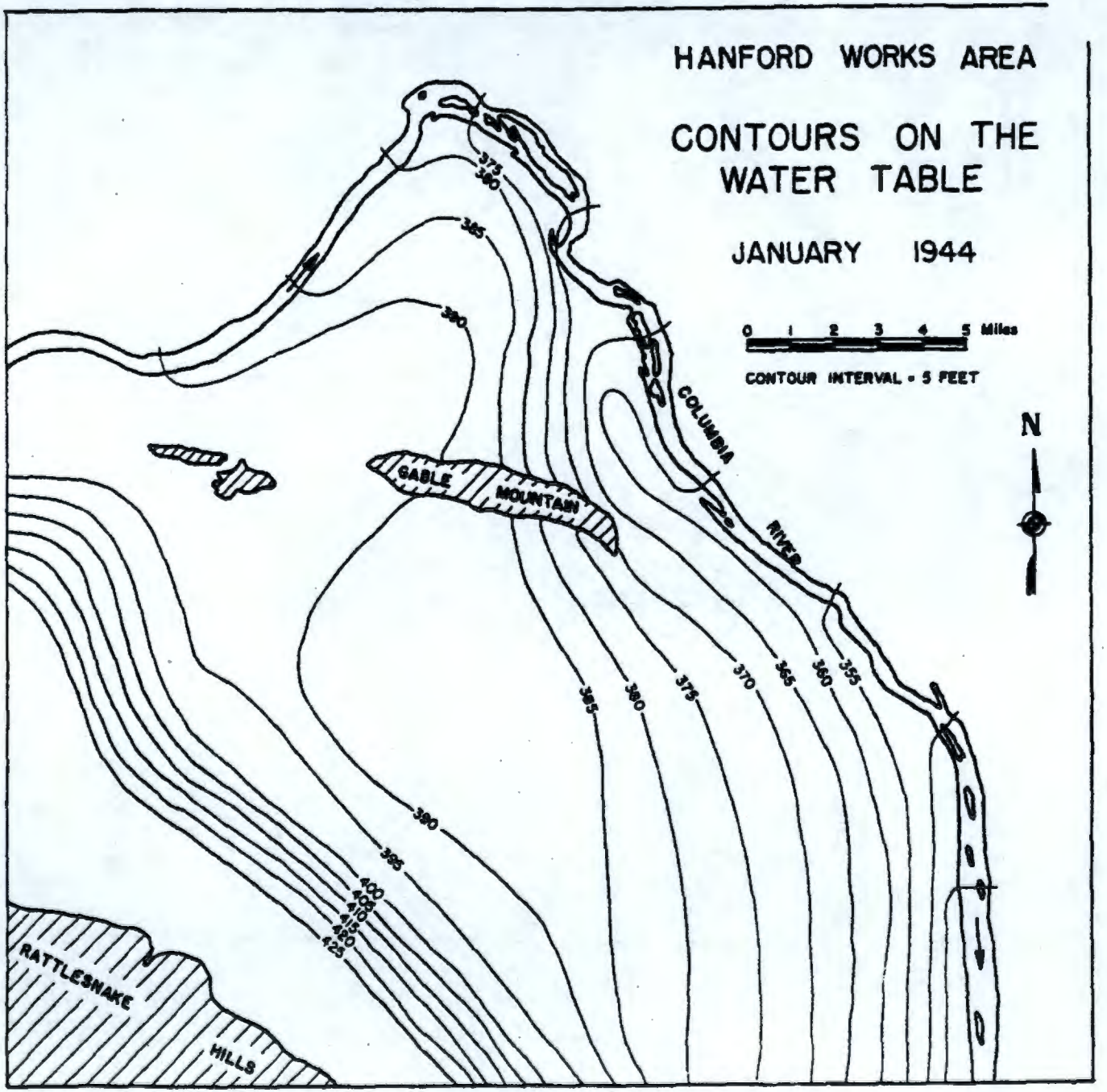

HANFORD WORKS AREA
CONTOURS ON THE WATER TABLE
JANUARY 1944
0 1 2 3 4 5 Miles
CONTOUR INTERVAL = 5 FEET
N
COLUMBIA RIVER
GABLE MOUNTAIN
RATTLESNAKE HILLS
375
380
385
390
395
400
405
410
415
420
425
370
365
360
355

Figure 4

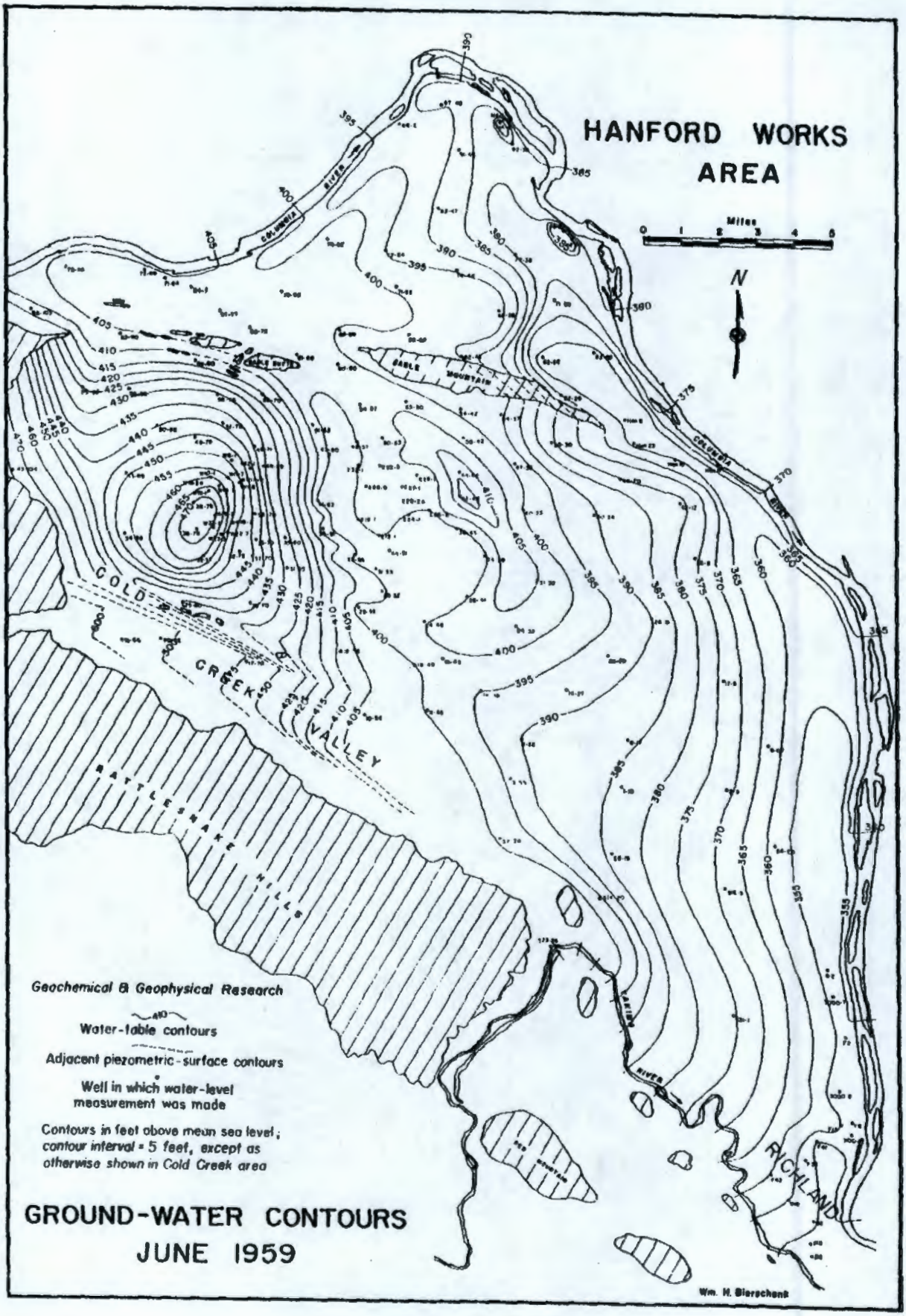

HANFORD WORKS
AREA
Miles
0 1 2 3 4 5
N
COLUMBIA RIVER
SABLE MOUNTAIN
COLD CREEK VALLEY
RATTLESNAKE HILLS
YAKIMA RIVER
RICHLAND
Geochemical & Geophysical Research
Water-table contours
Adjacent piezometric-surface contours
Well in which water-level measurement was made
Contours in feet above mean sea level; contour interval = 5 feet, except as otherwise shown in Cold Creek area
GROUND-WATER CONTOURS
JUNE 1959
Wm. H. Bierschenk

Figure 5

water table but also to depths below the water table and generally upward again to the water table at some other place.

As Figure 5 indicates, the pattern of ground-water movement underlying the Hanford plant has changed fundamentally during the 14 years of plant operation, owing to concurrent changes in water-table form. In brief, the zone saturated by infiltered waste effluents creates a ground-water divide between two well defined mounds with intermediate-level waste disposal sites located on the inner flanks of the mounds and the southern slope of the saddle between them. From the northern or outer flank of this system, the artificially recharged water largely moves radially northwestward and northeastward. From the southern or inner flank of the divide the infiltered wastes converge and move generally southeastward swinging eastward as their travel path lengthens (1).

The directions of movement described above are those which would be taken currently by any radioactive waste products infiltering to the water table from an overlying disposal site. It must be recognized, however, that deposits of sand and gravel such as comprise the glaciofluviatile and Ringold sediments are, in varying degree, lenticular, and in coarse lenses wastes would move more rapidly than in the fine materials. In addition, if the lenses are elongated in one direction or the strata are inclined steeply, the direction of flow will incline in the direction in which water moves most easily. Furthermore, a waste contained in a stream of ground water disperses both along and transverse to the direction of flow. As pointed out by Theis (31), dispersal in the direction of flow reduces the concentration of the contaminant if the waste is a slug temporarily introduced and gives a warning at a locality downstream of the approach of a continuously introduced waste stream. Dispersal across the direction of flow spreads a contaminant more widely but reduces the concentration.

Such factors as heterogeneity, anisotropy, and dispersal assume great importance in determining the path of contaminants in the ground water. Consequently, the estimated mean lateral path of ground-water contamination derived from water table contour maps is taken to represent the probable minimum distance of travel from beneath disposal sites to the Columbia River. Based on the hydrologic conditions inferred from Figure 5, a minimum path from the disposal sites to the river of from 29 to 34 kilometers appears reasonable.

The rate of ground-water flow is fixed by the vector quantity describing the maximum hydraulic gradient. Darcy's law for laminar flow is applicable but enables the estimation of only average velocities. Variation from the average is likely to be considerable so that some small fraction of the flow may move at several times the average velocity. For example, fluorescein tracers have been detected in observation wells at various distances down gradient from injection wells. Rates of travel of the dye, based on

the first detected arrival, have been measured to be 52 meters/day through 15 meters of travel in one case; 52 meters/day through 3,500 meters and 60 meters/day through 4,100 meters in a second case; and 135 meters/day through 2,700 meters in a third case. These velocities are 3 to 4 times greater than the calculated average values.

Along the estimated hydraulic path from ground disposal sites to the river, ground-water movement occurs under an average hydraulic gradient of about 0.004 in the Ringold aquifer of permeability estimated to be 1.4×10^{-2} cc/sec/cm^2. The effective porosity of this aquifer is estimated to be 10%, therefore the average rates of movement are calculated to be about 0.5 meters/day. Movement in the highly permeable glaciofluviatile sediments occurs chiefly under shallow gradients of only about 0.0001 and the average permeability is assumed to be 2.5 cc/cm^2/sec. Average velocities of about 2 meters/day are calculated for this portion of the path assuming an effective porosity of 10%.

Direct movement to the Columbia River through glaciofluviatile sediments is inhibited by the relative positions of the ground-water mounds. Instead, general movement occurs more to the south through Ringold deposits. Based on the average ground-water velocities calculated from measured gradients and estimated permeabilities and porosities of the aquifers affected, a "travel time" of about 180 years is calculated for ground-water flow from ground disposal sites to the river. It is recognized however that the maximum rate of movement of the ground water and even of some materials dissolved in it (e.g., ruthenium-106 and nitrates) may be several times the average. On the other hand, those dissolved constituents that enter into adsorption reactions (e.g., strontium-90 and cesium-137) will move far slower than the water, the chemical nature of each ion establishing the degree of retardation. Consequently, "isotope travel time" is suggested as a more descriptive term for the actual occurrence of concern, the ground-water movement rate being of significance as it represents a maximum conceivable isotope travel rate.

Studies at the University of California Sanitary Engineering Research Laboratory have shown that hydraulic phenomena produce velocity variations that bring about a longitudinal mixing of selected intruding and displaced fluids. A diffuse zone or "concentration front" forms rather than a sharply defined interface (23) (27). The depth of this zone increases in proportion to the distance traveled due to portions of the intruding contaminant moving at velocities exceeding the average. They also point out that ion exchange reactions may modify the propagation of a radiocontaminant in two ways: 1) the median velocity of the contaminant front will be predictably less than that of the liquid front, and 2) the depth or diffuseness of the front may be modified over that resulting from purely hydraulic phenomena. When the radiocontaminant is not selectively sorbed by the exchange medium, the front will become increasingly diffuse as it progresses through the medium. When the radiocontaminant has a selective affinity for the medium, as may be the case with strontium or cesium as the displacing cation, the front may rather tend to sharpen as propagation continues.

Empirical data obtained from radiological monitoring of wells at Hanford have shown that the chemical form of Ru^{106} in Hanford wastes prevents this isotope from being significantly affected by ion exchange, and anionic components of waste such as nitrates are apparently not affected at all. Much additional study is needed before the existing knowledge of ground-water travel rates can be applied to the calculation of isotope travel rates for all of the constituents of interest in radioactive wastes.

Another factor requiring study is the "sinking" of wastes. It is recognized that the possibility exists for high density wastes to settle in the zone of saturation by gravity. This concept presumes that the vertical movement under gravity will be significant compared to horizontal movement rates and with rates of dispersion throughout the aquifer. To date, no clear cut evidence of this phenomenon has been obtained in field studies of waste movement. In general, irregularities in the contamination pattern observed are explainable on the basis of formation inhomogeneity and anisotropy, but a more systematic study of the phenomenon is contemplated.

CONCLUSIONS

Although low concentrations of various waste components have been found in wells which monitor the ground waters near disposal sites, ground waste disposal practice at Hanford has been so managed that no movement of fission products to the river has been detected. The radioactive material in the Columbia River is monitored isotopically at the points where river water is used, and their exposure potential is summed to permit their evaluation (20). All radioactive material in the river except that naturally present may be presumed to originate from reactor effluent.

The calculated average exposure to persons near the plant as a result of drinking water from the Columbia River is less than 20% of the maximum permissible limit for members of the public living in the neighborhood of controlled areas and the exposure received by persons from consistently eating some kinds of fish caught near the plant can amount to about this same fraction. Swimming or boating on the river and the irrigation of crops provide only minor contributions to the total exposure. No restrictions on downstream use of the river have resulted from the presence of the radioactive effluent.

It has thus been illustrated that, for an environment of the Hanford type, low-level radioactive waste disposal to a river and simultaneously to the ground in its neighborhood may be practiced with safety. This "dilute and disperse" waste disposal policy is a sound method of dealing with the large volumes of low and intermediate level wastes that necessarily are generated by nuclear plants. Hanford experience illustrates a method of taking advantage of natural environmental features in optimizing plant operation. A clear understanding of the many factors involved in dealing with waste disposal problems is, in fact, necessary to wisely select a site for such an installation.

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