Atomic Energy of Canada Limited

MOVEMENT OF RADIOACTIVE WASTE THROUGH SOIL
5. The Liquid Disposal Area

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by

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Chalk River, Ontario

June, 1962

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SYNOPSIS

Two seepage pits, used for the routine disposal of water containing low-level radioactive wastes, have been examined by a soil and groundwater survey.

Reactor Pit 2 has received 1.75×10^8 gallons and has absorbed 11,000 curies of soluble β-emitting radionuclides with 90g Pu. Of these, 87 curies have migrated as cations and the weak 'front' of this movement, containing Sr-90, Co-60, Cs-137 and Ce-144, is expected to be released into the runoff of a nearby swamp in 4 years.

The Chemical Pit, used for the disposal of low-level moderately acid waste has accumulated 500 C of total β and 50g Pu. It recently released Sr-90 and Co-60 into a nearby swamp three months after receiving a disposal containing high concentrations of complexing agents. Soil and groundwater sampling have shown that the ion-exchange capacity of the soil has been greatly reduced and that the escape of Co-60 will increase to roughly three times its present value. The escape of Sr-90 is not expected to increase.

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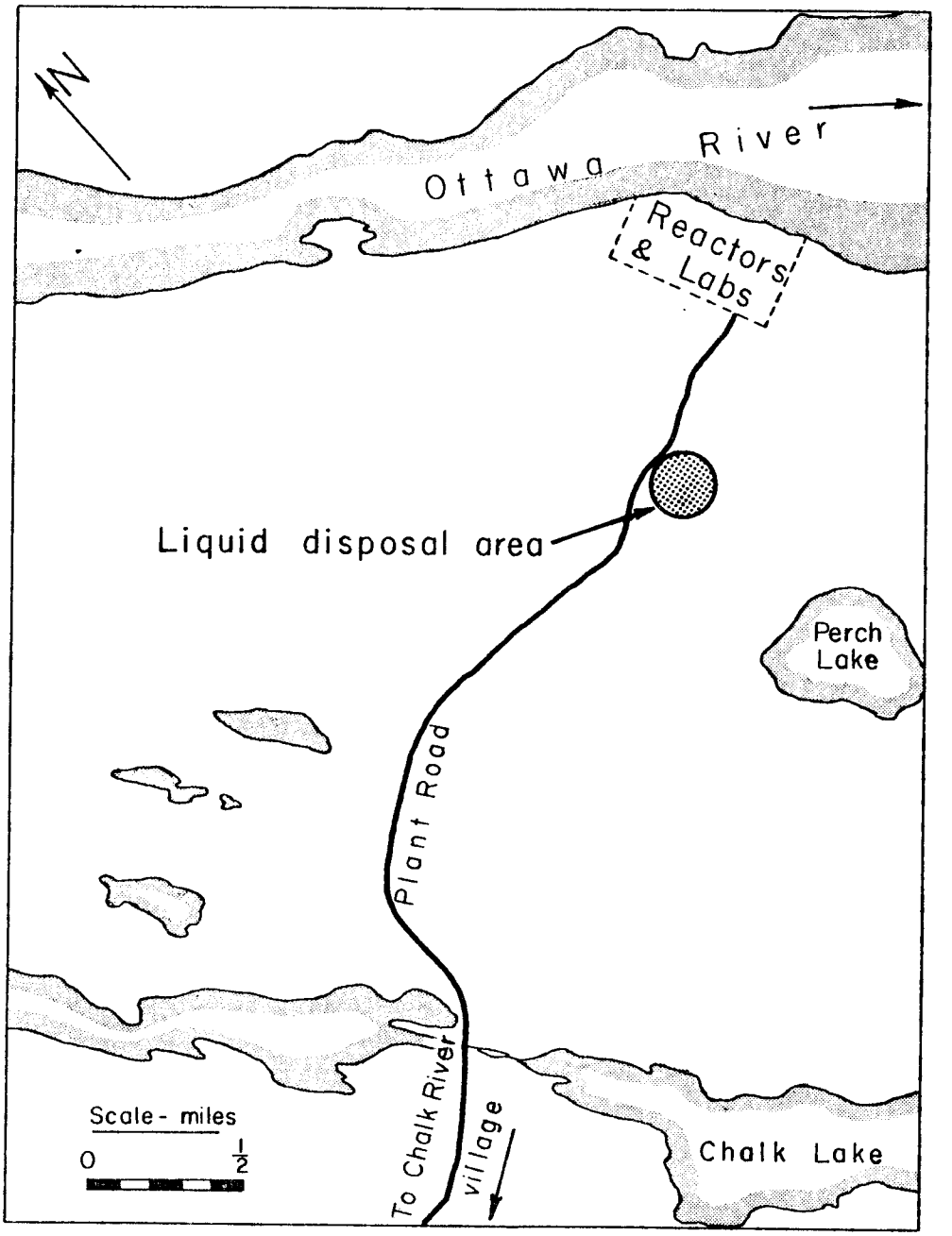

Site of Liquid Disposal Area within the Outer Area of the Project.

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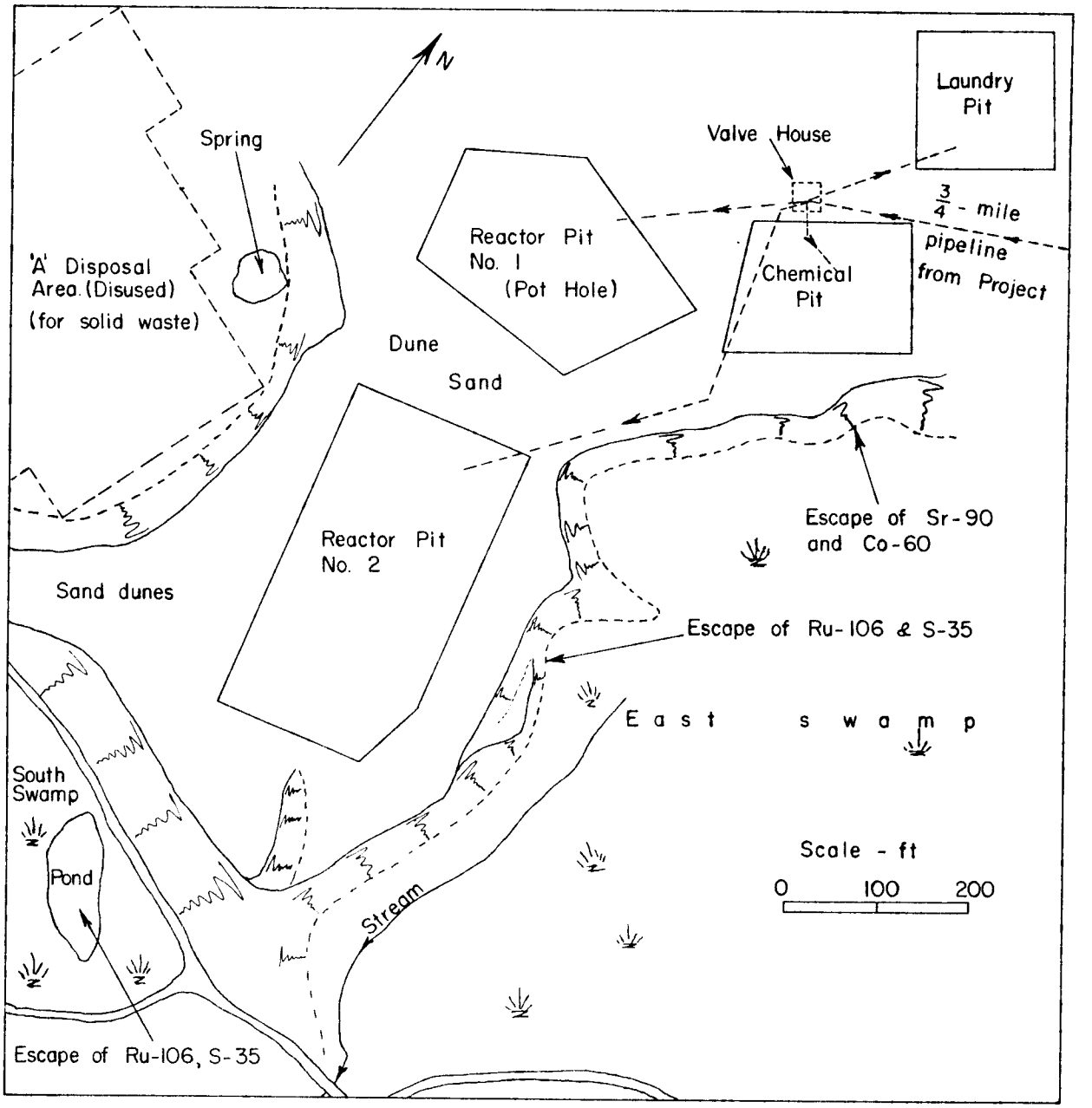

General Plan of Liquid Disposal Area. 3/4 mile from Plant Area.

INTRODUCTION

Special seepage pits in the Liquid Disposal Area at the Chalk River Project receive all the waste water of low activity that is unsuitable for discharge directly into the Ottawa River. The porous nature of the soil allows the water to percolate into the natural groundwater system where many of the hazardous radionuclides are absorbed by the soil. This practice was started in 1953 when a 3/4-mile pipeline was constructed to carry waste water from the establishment to a suitable area of dune sand where it was discharged into a natural pot-hole. After three years and a mean daily discharge of 50,000 gallons, vegetation growing in the pit became radioactive and radiostrontium appeared in the seepage from a nearby spring. In 1956, use of the pothole was discontinued; it was covered with a layer of uncontaminated soil and the water was diverted into three new pits that were to form the liquid disposal area. (General Plan). Each of these was reserved for a special type of waste; Reactor Pit 2 for large volumes of water from the spent fuel storage bays, the Chemical Pit for solutions of variable pH and chemical composition and the Laundry Pit for water from the active laundry. The Laundry Pit did not accumulate a significant amount of radionuclides and it has not been used since 1958. However, Reactor Pit 2 and the Chemical Pit continue to absorb a large daily effluent from the Project and escapes of radionuclides into the neighbouring East Swamp have undoubtedly originated from these two pits.

An examination of the soil surrounding the original pot-hole and the pits has been carried out in order to find the extent of radioactive migration underground and to determine whether the present escape portends an imminent large-scale release of radioactivity.

PROCEDURE

The soils of the liquid disposal area were examined by drilling several holes and collecting undisturbed soil samples that were tested for permeability and grading. The casings were forced into the soil by washboring and soil samples were collected with a compressed-air sampler[1]. It was not possible to penetrate the deeper deposits of glacial till by this method because progress was halted by boulders. However, earlier borings had been carried out with a churn drill in order to install monitoring tubes down to bedrock and the results of both series of drillings were used to determine the contours of bedrock and glacial till.

Groundwater elevations were measured by installing a standpipe in each borehole. Where further data were needed to plot detailed variations in water table contours, temporary standpipes were driven con-

sisting of E drill rods and hollow brass drive points with sidewalls of porous bronze[2]. Since the internal diameter of the rod couplings was only 0.43 in., the depth to groundwater was measured with a probe (1/8" dia) that made electrical contact with the water surface.

A more detailed soil investigation followed, in which a multiple soil sampler[3] was used to collect large numbers of saturated soil samples, suitable for radioassay or radiochemical analysis. In a single drive of this apparatus soil samples could be retrieved from various horizons down to the limit of penetration. Normally, samples were taken at 2-ft intervals in depth.

Sampling was started close to the edge of each pit and continued at increasing distance until "inactive" soil was encountered; the extent of migration was thus spanned and the "front" delineated. Soil samples were examined by counting 50 mg of dry soil for total β activity in a β counter. A gamma spectrometer was used to identify the γ-emitting radionuclides in the samples.

In order to compare the relative contamination of the soil with that of its accompanying groundwater, samples of groundwater were collected from identical positions where soil had previously been sampled. Each water sampler was a modified porous bronze piezometer[2] (1 ft in length), connected as a drive point to the toe of a string of 'E' drill rods. Groundwater entered through the bronze sidewalls into the inner chamber (100 ml) and was withdrawn to the surface through twin polyethylene tubes within the hollow core of the drill rods. An evacuated flask connected to the top of one tube would remove the entire sample while the other tube vented the sampling chamber.

The rate of groundwater movement and its direction were measured by injecting fluorescein into the groundwater and sampling it from positions "downstream". Both the injection and the subsequent dye sampling were carried out with porous bronze samplers.

RESULTS

The soil survey showed that the sand dunes in the liquid disposal area were 25 - 35 ft thick. Wind-blown sand near the surface overlaid water-deposited sand that was laminated horizontally. At Reactor Pits 1 and 2 about 10 ft of glacial till lay beneath the sand and formed an impermeable medium over the bedrock. At the Chemical Pit, however, a 6-ft deposit of silt lay between the sand and the underlying glacial till.

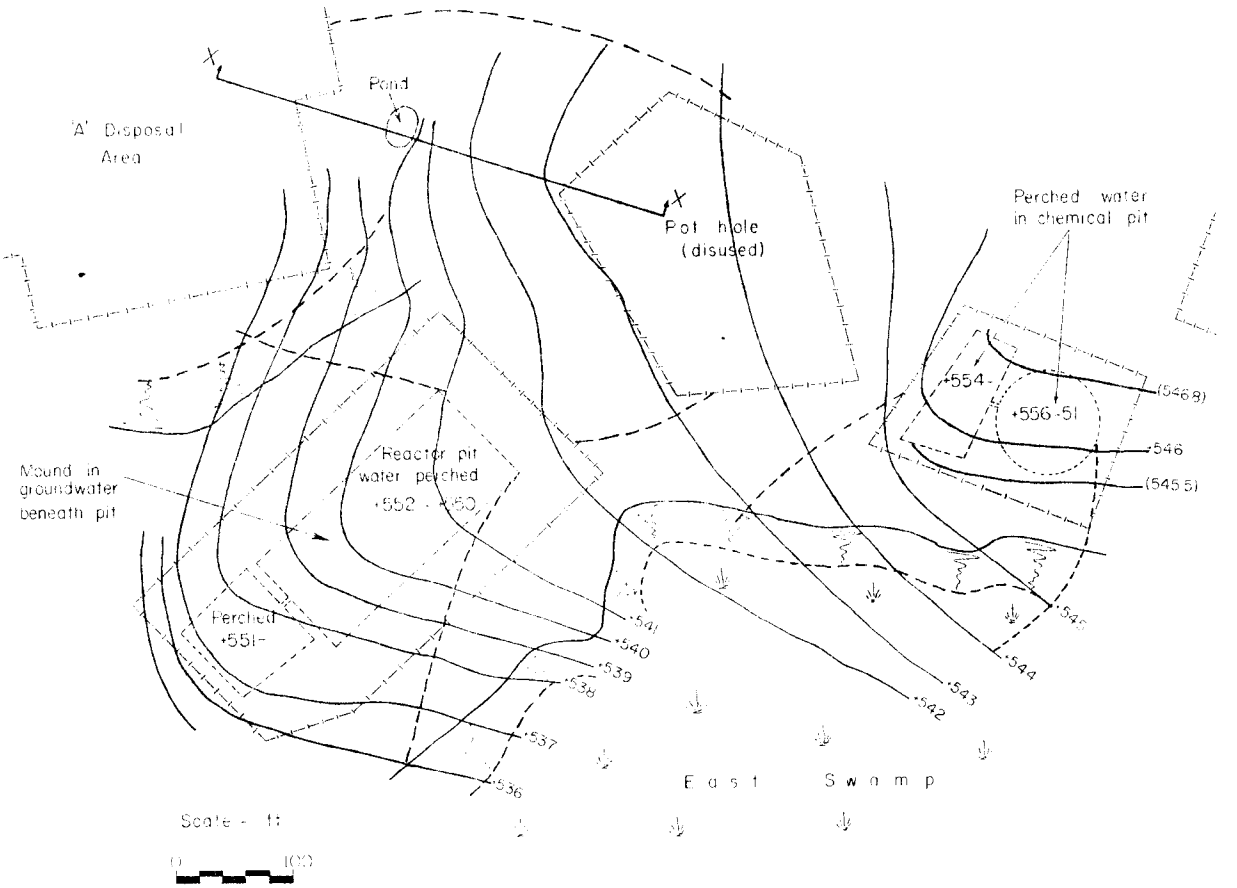

Fig. 1 - Contours of water table beneath Liquid Disposal Area. Shaded areas show possible regions of migration from each facility. XX is the cross section shown in Fig. 2.

Water Table

Contours of the water table are drawn in Fig. 1. The groundwater surface lay in the sand deposits and the anticipated direction of migration through the sand has been shown by shading for each pit. Migration from Reactor Pit 1 (pothole) lay towards 'A' disposal area and Reactor Pit 2; from the Chemical Pit it was towards East Swamp. A slight mound in the groundwater beneath Reactor Pit 2, caused by recharge from the pit, has produced a radial migration to fan from three sides of the pit.

Reactor Pit 1 (Pothole)

This was the first site used for the routine disposal of radioactive water and it consisted of a natural depression in the sand dunes of roughly 150 ft diameter. The pothole was used from 1953-56, receiving an estimated[4] 2000 C of Sr-90 plus many other fission products, and 100 g of Pu. The first escape of fission products was noted four months after disposals began, when ruthenium-106 appeared[5] in a pond 300 ft to the south-east. A longitudinal section between the pit and this pond (Fig. 2) shows the migration along this path. The convergence of the groundwater surface with the rising profile of glacial till caused seepage in the form of a spring at the till outcrop. This has produced a pond and the overflow from this returns below ground into the sands of the 'A' Disposal Area.

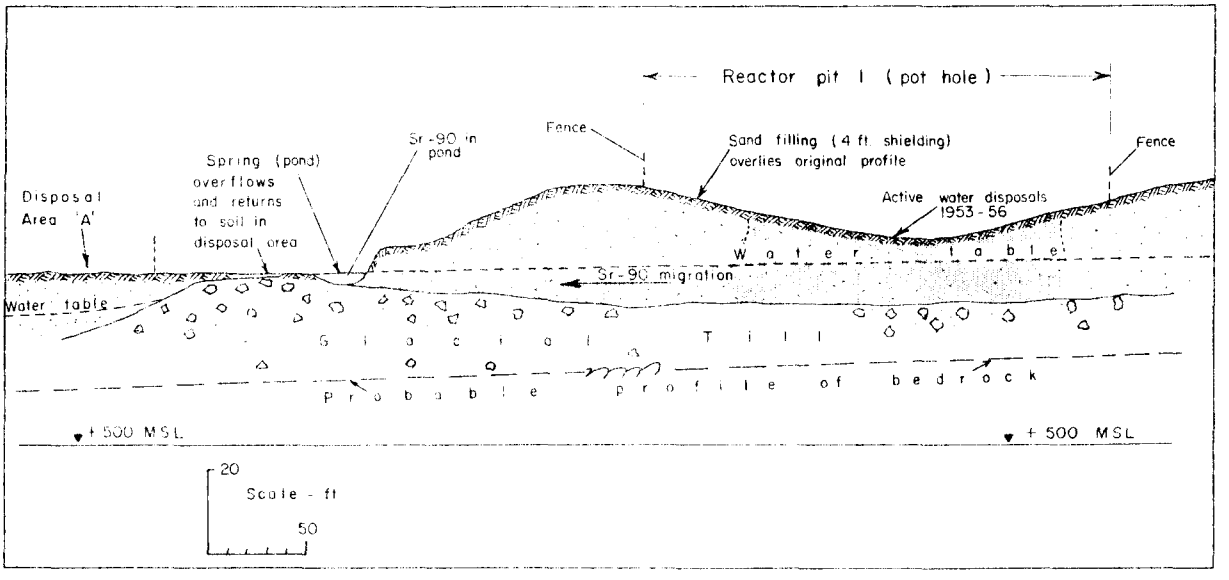

Fig. 2 - Longitudinal section between Reactor Pit 1 and the contaminated spring. The shaded area shows the migration of Sr-90 in the groundwater.

Further soil sampling confirmed that Sr-90 has migrated in the soil towards Reactor Pit 2 and it is probable that some has penetrated beneath the pit. Other soil samples between the pothole and East Swamp have shown that the boundary of low-level contamination lies 80 ft from the swamp. This position is outside the range of movement that would be anticipated from the present water table contours. It is probable, therefore, that when the effluent was routinely discharged into the pothole, a mound formed in the groundwater, similar to the one beneath Reactor Pit 2. This produced a broader range of possible migration. The results confirm that no radiostrontium from the pothole ever reached East Swamp.

Reactor Pit 2

Reactor Pit 2 is a pebble-filled pit excavated in dune sand. It is 8 ft deep with sloping sidewalls and has an estimated capacity of 4.5×10^5 gallons. When it came into operation in 1956, the volume of water pumped into it averaged 1.6×10^6 gals/month, but this flow had increased to 4×10^6 gals/month by 1960. The pit was unable to receive this additional flow and the water level rose to within 2 ft of the top. In August 1960, therefore, an auxiliary overflow pit of the same type was added at one end and connected to the main pit by an overflow sill. After April 1961, the volume of influent was reduced following the introduction of a recirculating-water treatment plant for rod-bay water. Thus the combination of reduced input and the new overflow pit lowered the depth of standing water to about 2 ft.

Between 1956 and December 1961 the pit absorbed 175×10^6 gallons containing an estimated 11,000 nominal curies of soluble radionuclides.

A few months after the pit was put into service, an escape of ruthenium-106 and sulfur-35 was found in the surface water of East Swamp, 200 ft from the pit. Later (1958) these radionuclides were recognised in the surface waters of South Swamp 300 ft away and in a sampling well 600 ft to the west[6]. These releases have continued and are evidence of the wide-angle of migration as inferred from the water table contours. Both ruthenium-106 and sulfur-35 are in anionic form, and they travel at almost the same rate as the groundwater. The latest[6] rate of release of S-35, the principal contaminant into East Swamp, is 75 mC/month.

A plan of the boreholes driven around Reactor Pit 2 is shown in Figure 3. The limiting boundary or "front" of radioactive cations is also plotted showing the principal migrations to the east and west. Migration to the south also developed between 1956-1960 but it is concealed beneath the auxiliary pit; subsequent migration from the new boundary has been small.

The cations of this migration are strontium-90, cobalt-60, cerium-144 and cesium-137 and at present, on the east side, their "fronts" are coincident; the relative migration rates might well make them diverge later so that individual 'fronts' may be distinguished.

Boreholes were sunk on lines normal to the edge of the pit, the first sampling points being at the top of the sloping sidewall at the junction of the pebble fill and sand. Sampling was continued at intervals along

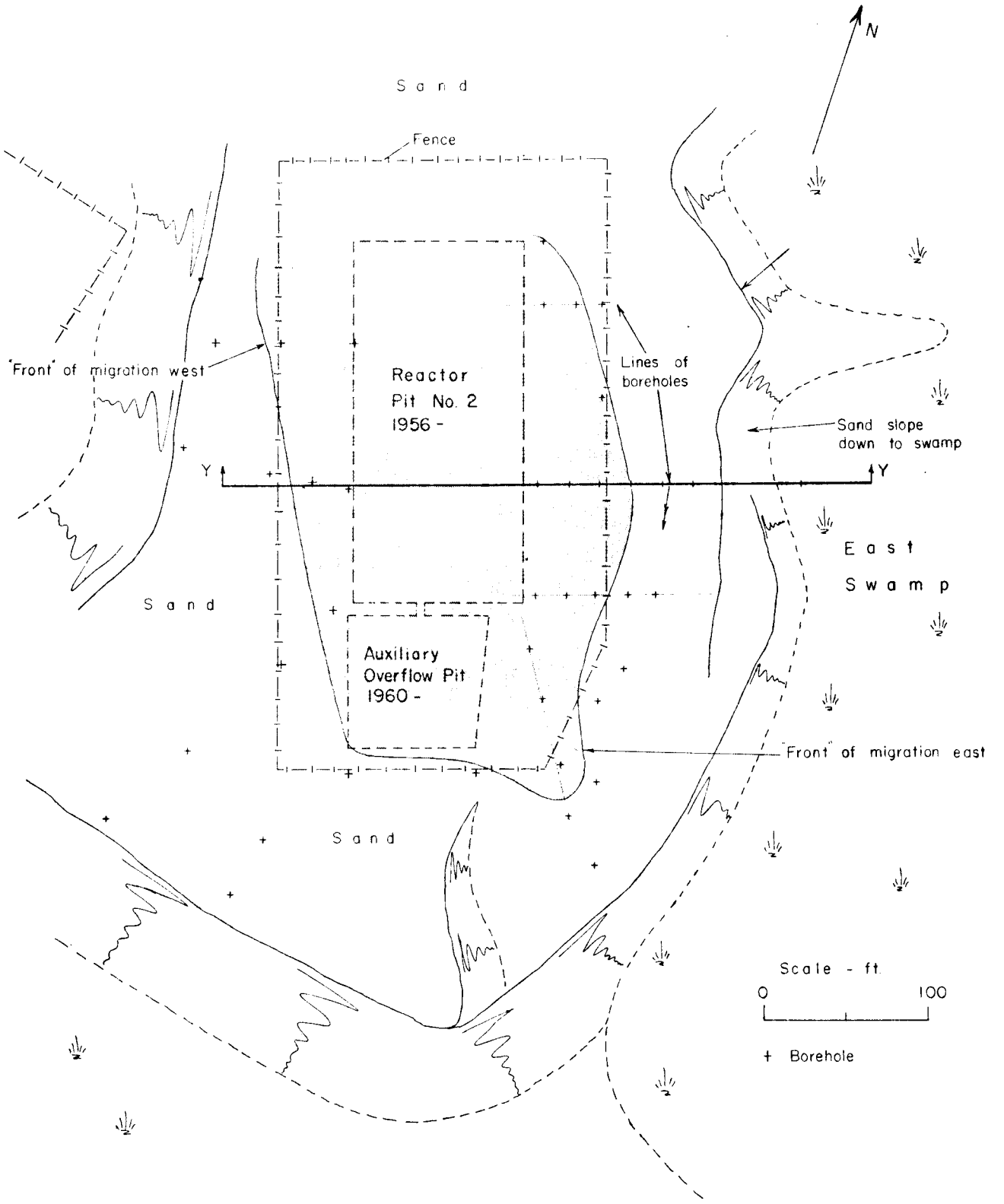

Fig. 3 - Plan of borings near Reactor Pit 2. The area of present migration of cations is shaded. YY is cross section shown in Fig. 4.

each line until the track of migration had been spanned. On both the east and west sides the quantities of migrant fission products in the soil have been estimated from counts on the soil samples and the natural porosity of the soil. Estimates of the nominal curies in vertical sections were made at 0, 18, 36 and 54 ft from the pit edge in order to compute the number of nominal curies in each migration.

These results, summarised in Table I, show that about 50 curies have moved towards East Swamp and 37 curies have moved westwards towards the 'A' Disposal Area.

TABLE 1

Migration From Reactor Pit 2

Distance of section from pit ft	Fission product concentration: At section Curie/ft	Fission product concentration: Mean between sections Curie/ft	Nominal curies between sections	Total nominal curies in migration
	Migration East			
0	2.67			
		1.925	34.6	
18	1.18			
		0.73	13.1	
36	0.28			
		0.14	2.5	50.2
54	0			
	Migration West			
0	0.97			
		0.67	30.0	
45	0.37			
		0.185	7.4	37.4
85	0			

Water samples were collected from a number of boreholes to investigate the Distribution Ratio of the radionuclides. The Distribution Ratio (D) is the ratio of activity on the soil to the activity in the water within a volume of undisturbed soil; in this case the value refers to a composite of the radionuclides. The values of D varied from 50 - 250 in boreholes between the pit and the swamp. Similar values were measured in South Swamp where D = 60 for high concentrations of Sr-90 and south

of A Disposal Area where D = 140 - 300 for migrations of mixed fission products[7]. Thus the ion-exchange properties of the soil near Reactor Pit 2 are similar for both filtered pit-water and groundwater.

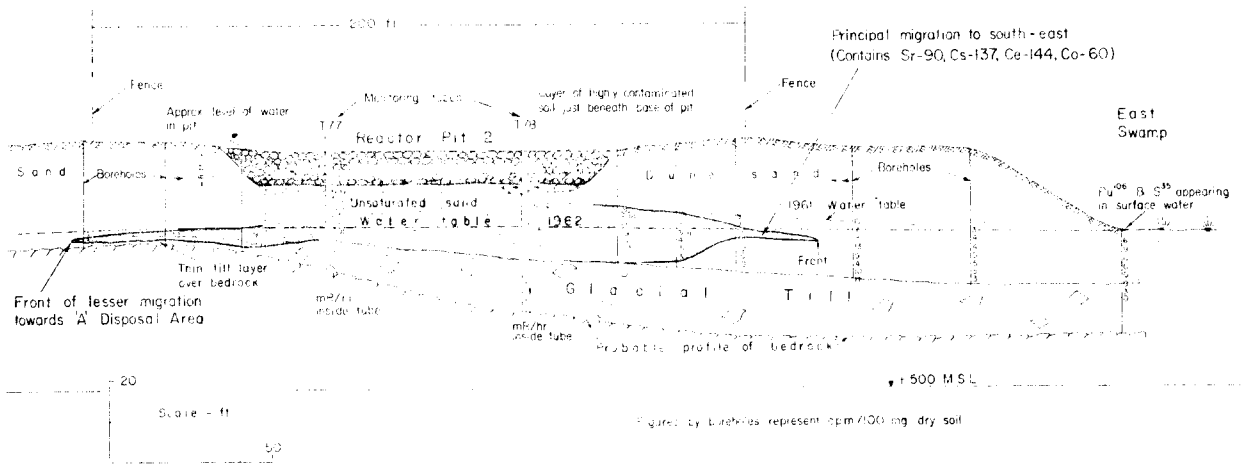

Fig. 4 - A West-East cross section through Reactor Pit 2. The shaded area shows migration from either side of the pit and the zone of high contamination at the pit base.

A west-east cross section through the pit is drawn in Fig. 4, showing the two migrations in opposite directions. The glacial till and underlying bedrock rise to a ridge near the western edge of the pit and as a result the aquifer to the west is only 4 ft deep while the eastern counterpart is 10 ft or more. The tracks of radionuclides extend through the entire thickness of saturated sand but have not penetrated the glacial till. The higher concentration of contaminants lies in the eastern migration and because this is potentially more hazardous in its approach to the surface waters of East Swamp, it has been examined in greater detail.

Figure 4 shows that the water table (in 1962) lies 10 ft below the base of the pit while 2-3 ft of free or "perched" water remains in the pit. The reduced permeability of the sands lining the pit has probably been caused by an accumulation of sludge (diatomaceous earth) that was occasionally pumped in as a slurry following a backwash of filters in the storage-bay system where the disposals originated.

Two aluminum monitoring tubes extending down to bedrock, were installed in the pit. Radiation measurements inside these confirmed that there was no contamination in the deposits of glacial till and that the highest radiation field lay close to the base of the pit and in a narrow band of unsaturated sand beneath it. (Heavily shaded zone in Fig. 4)

The build-up of radiation from 1957 onwards was recorded by radiologging in these tubes. In 1958 radiation fields existed in a band near the pit base and over the next three years the thickness of this band increased upwards into the pit. It is probable therefore that most of the radioactivity at present fed to the pit is being absorbed on the sludge material with its high ion exchange capacity for cations.

The tracer test with fluorescein dye[2] showed that the velocity of groundwater near the present 'front' was 2.2 ft/day, measured at several sampling points along a total movement of 20 ft. Thus the time taken for groundwater to move from the pit to East Swamp, along the estimated 300-ft inclined path, is about 4.5 months. Previous measurements[7] of the rate of Strontium-90 movement (0.027 x vel. of groundwater) in this area, showed that the present (1962) 'front' of strontium is likely to traverse the remaining 90 ft to the East Swamp in $\frac{90}{2.2} \times \frac{1}{0.027} \times \frac{1}{365} = 4.1$ years

The 'front' of migration to the west is farther from the pit than the eastern front since it is moving faster. However, owing to the shallow aquifer, the number of curies is smaller and the migration is less important because it will remain below ground in its passage through the 'A' Disposal Area, where numerous highly active disposals are sources for other independent migrations[7].

Chemical Pit

The chemical pit is a circular (88 ft dia) pebble-filled pit similar to Reactor Pit 2 but with a smaller capacity of 1×10^5 gallons. It was first used in March 1956, and after one year's satisfactory operation the rate of seepage diminished and the level of the standing water rose in the pit. An auxiliary overflow pit was therefore built in 1958. The pit normally receives about 3×10^5 gals/month of moderately acid liquid waste (pH 3-5), originating mainly from laboratories, and by the end of 1961 the accumulation of fission products in the pit had reached an estimated 466 curies.

In September 1961 there was an unusual liquid disposal containing citrate and possibly other complexing agents. In January 1962, routine sampling[6] of water issuing from the East Swamp showed a large increase in the concentration of Sr-90, accompanied by detectable quantities of

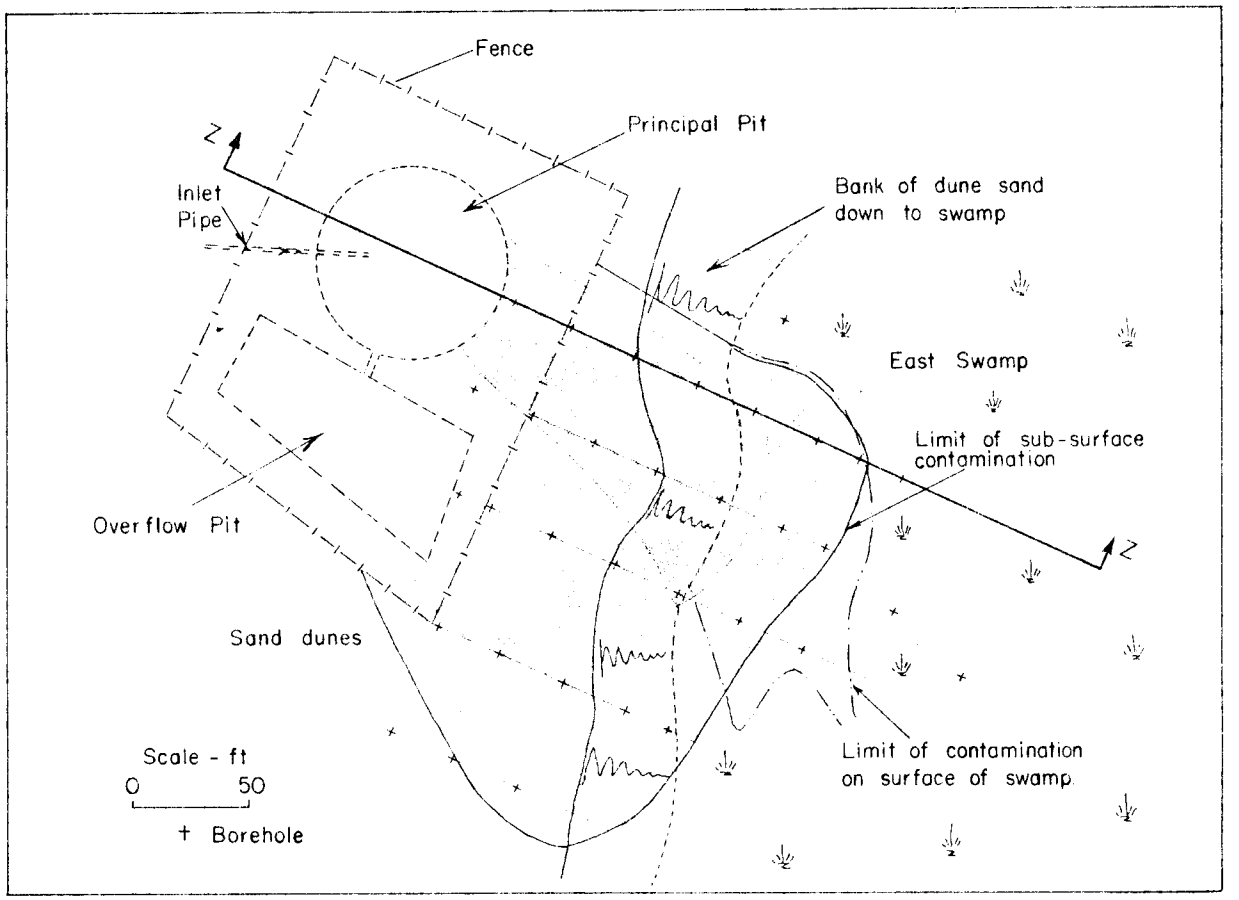

Fig. 5 - Plan of Chemical Pit showing layout of borings, shaded areas show limits of migration - the principal escape is in dense shading. ZZ is the line of the cross section in Fig. 6.

cobalt-60. An area of East Swamp close to the chemical pit was found to be radioactive and a more detailed examination delineated a 60-ft x 30-ft surface area contaminated with cobalt-60. (Fig. 5). This region lay on the edge of the swamp closest to the chemical pit.

The soil and groundwater were examined by drilling at 30-ft intervals in the dune sand between the pit and the swamp. Soil and water samples were collected every two feet down to a depth where no further activity occurred.

The plan of the borings and the outline of the migration in 1962 are drawn in Fig. 5, showing that the principal migration was from the origi-

nal (circular) pit. The direction and boundaries of the migration agree with those anticipated from water-table contours. Surface contamination of the swamp has also been plotted.

Radiochemical analyses showed that, in general, only two radionuclides were present viz. Co-60 and Sr-90. The relative counts on soil and groundwater samples from the same position showed that frequently the ratio $\frac{\text{CPM/100 mg Soil}}{\text{CPM/ml Water}}$ was an order of magnitude lower than at other places in the disposal areas e.g. Reactor Pit 2. Beacause of this high concentration of contaminants in solution, the groundwater samples were examined to assess the relative proportions of Co-60 and Sr-90.

A 10-ml portion was evaporated from each sample, counted in a gamma spectrometer to estimate the cobalt present, and then recounted in a β counter to measure the β particles from both radionuclides. After calculating the counting efficiencies of each instrument for both Co-60 and Sr-90, the strontium content in each sample was estimated by difference. A similar test was carried out on some of the soil samples, showing that the $\frac{\text{Sr-90}}{\text{Co-60}}$ ratio on the soil was roughly the same as in the associated water samples.

An estimate of the size of the migration was made by calculating the number of curies in vertical parallel sections at 0, 30, 60, 90, and 120 ft from the pit and the contamination in each section (in curies/ft) is shown in Table 2.

TABLE 2

Contamination of Soil and Water by Co-60 and Sr-90 from Chemical Pit

Distance from edge of pit ft	SOIL Conc. of Sr-90 + Co-60 Curies/ft	WATER	
		Conc. of Sr-90 Curies/ft	Conc. of Co-60 Curies/ft
0	0.266	0.0091	0.0257
30	0.260	0.0113	0.0108
60	0.296	0.0269	0.0408
90	0.290	0.0197	0.0176
120	0.140	0.0266	0.0129
Break through in swamp	+(0.106 humus)		

A summation of these results shows that 32 curies (Co-60 + Sr-90) have migrated into the soil outside the chemical pit and that the associated groundwater contains 2.22 curies Sr-90 and 2.56 curies Co-60. The mean distribution ratio D for the combined radionuclides is $\frac{32}{4.78} = 6.7$, an order of magnitude lower than that for the migration from Reactor Pit 2.

This loss of ion-exchange capacity by the soil is attributed to the low pH and high-ionic content of the solutions discharged into the pit.

The figures in Table 2 show that the concentration of Sr-90 in the groundwater does not vary appreciably along the migration path between pit and swamp. However, the concentration of cobalt does vary, reaching a maximum half way along the path. It is anticipated therefore, that the rate of release of cobalt-60 into East Swamp will rise to roughly three times its present value as the peak of the migration approaches the swamp. The release should subsequently decline.

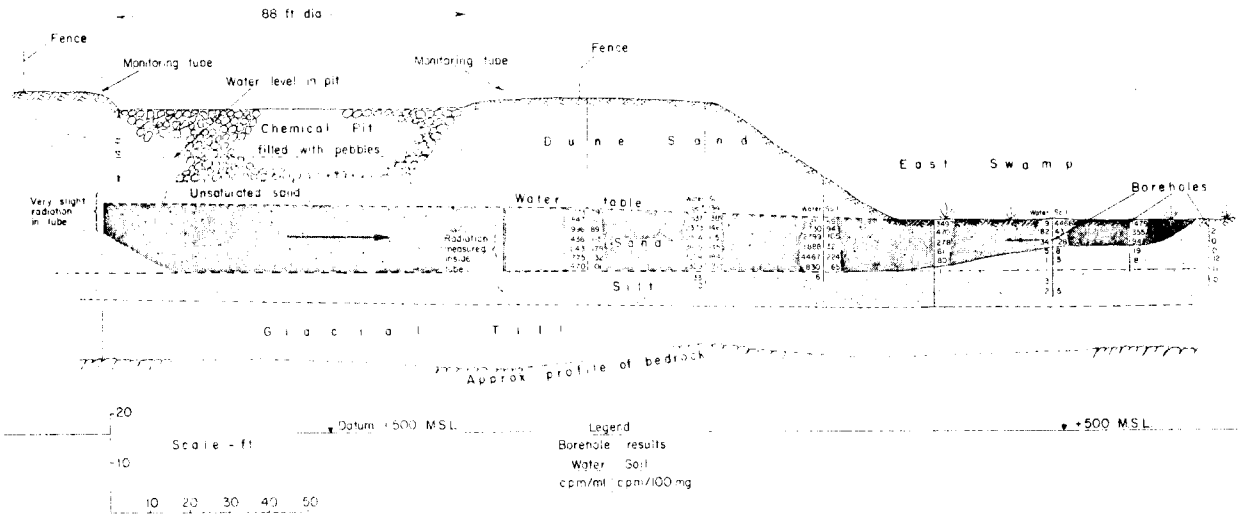

Fig. 6 - Cross section between Chemical Pit and Swamp. (ZZ on Fig. 5)

A cross section of the migration is shown in Fig. 6. This shows that bedrock is covered with glacial till and silt that have not been contaminated. The migration was confined to the saturated zone of sand, about 14 ft thick, overlying the silt. The water table dipped gently towards East Swamp, passing about five feet beneath the base of the pit.

Recharge from the pit caused slight mounding but not sufficient to promote radial percolation; thus the movement remained towards East Swamp. Free water in the pit was perched above a zone of unsaturated sand, duplicating the conditions found beneath Reactor Pit 2.

The surface of the East Swamp was composed of organic humus to a depth of about 2 ft. Fig. 6 indicates that the escape of radionuclides extended 75 ft into the swamp and humus samples in this region yielded high counts. This material has a much higher exchange capacity for cations than sand, so that high counts can accumulate from only moderately active seepage.

Other radionuclides found were ruthenium-106 in isolated pockets beneath the dunes, and cesium-137 in a large patch of humus in the swamp. Since cesium is normally fixed on the soil and migrates only slowly in a continuous track from the source[7], its presence in the swamp further demonstrates the large reduction in exchange-capacity of the soil.

CONCLUSIONS

Migration from Reactor Pit 1 is confined to Sr-90 and is directed towards the A Disposal Area where there is no possibility of imminent release into surface waters.

In Reactor Pit 2 the freestanding water in the pit is "perched" over a zone of unsaturated sand, intermediate between the pit-base and the water table a few feet below. Most of the contaminants fed to the pit lie in a band at the top of this zone extending upwards into an accumulation of sludge that lines the base of the pit. The pit has absorbed 1.75×10^8 gallons of water, containing an estimated 11,000 curies of soluble radionuclides, of which 87 curies have migrated as cations beyond the pit boundary. Thirty seven curies have moved west 80 ft towards the 'A' Disposal Area. This is the faster moving migration but, since it is destined to remain below ground for a further 700 ft and its path is through a region of much larger independent disposals, it has not been examined in detail. Fifty curies have moved in the opposite direction towards East Swamp and their escape into surface water is expected to start in four years. This migration is composed of strontium-90, cesium-137, cobalt-60 and cerium-144. Because the concentrations of these nuclides are low near the 'front', it may be several years after the initial escape before concentrations in the swamp run-off show a considerable increase.

If the disposals to the pit were halted, the configuration of the water table would revert to normal, thereby retarding the migration and directing it more to the south in a longer sub-surface path.

The Chemical Pit released strontium-90, cobalt-60 and some cesium-137 into East Swamp a few months after a disposal containing complexing agents. The tests on soil and groundwater samples between pit and swamp have shown that the ion-exchange capacity of the soil has been reduced by an order of magnitude owing to the ionic content of the mildly acid percolating solutions.

Both Sr-90 and Co-60 are distributed through the sand between the pit and the swamp. The concentrations of each radionuclide along this path show that the present release rate of Sr-90 into East Swamp will continue and that the Co-60 release will probably rise to three times its present value. It is anticipated that the area of surface contamination in the swamp will increase as a broader 'front' of migration intersects the swamp. The cesium-137 is probably fixed in the humus without contributing contamination to the run-off from East Swamp.

ACKNOWLEDGEMENTS

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REFERENCES

1. Parsons, P.J., 1960 Movement of Radioactive Waste Through Soil
1. Soil and groundwater investigation in Perch Lake Basin AECL-1038

2. Merritt, W.F., 1962 Routine Measurement of Groundwater Velocity using S-35. Health Physics Vol. 8 pp 185-189

3. Parsons, P.J., 1961 "Multiple Soil Sampler" Am. Soc. Civil Eng. Vol. 87. S.M. 6

4. Mawson, C.A., 1956 Report on Waste Disposal System at Chalk River AECL-344

5. Ophel, I.L., & Fraser, C.D., 1957 The Chalk River Liquid Disposal Area CRHP-709

6. Ophel, I.L., Private Communication

7. Parsons, P.J., 1961 Movement of Radioactive Waste Through Soil
3. Investigating the migration of fission products from high-ionic liquids deposited in soil. AECL-1325