

MOVEMENT OF RADIOACTIVE WASTE THROUGH SOIL

III: INVESTIGATING THE MIGRATION OF FISSION PRODUCTS FROM HIGH-IONIC LIQUIDS DEPOSITED IN SOIL

CRER-1018

by

P.J. PARSONS

Chalk River Nuclear Laboratories

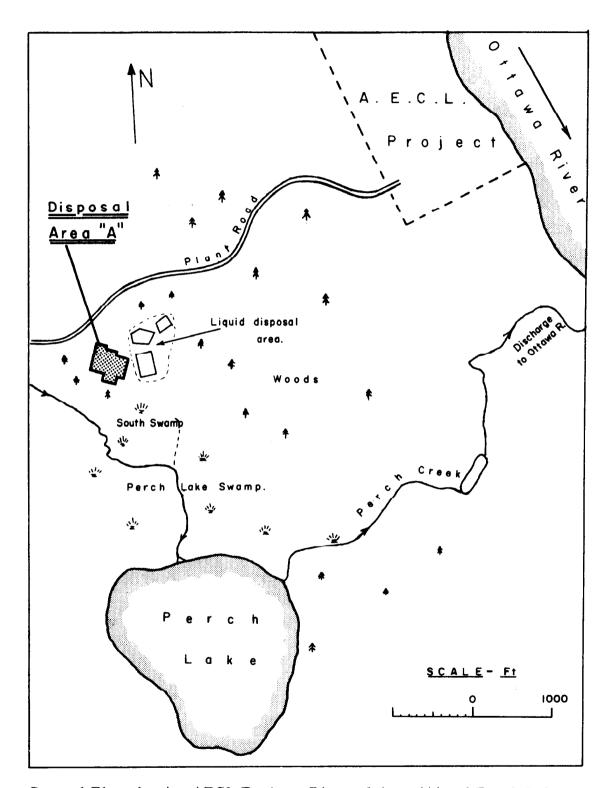
Chalk River, Ontario

August 1961

Reprinted March 1975

TABLE OF CONTENTS

	TABLE OF CONTENTS	Page
INTRO	DUCTION	1
INFOR	MATION AVAILABLE	1
PROC	EDURE	4
	Multiple soil sampler	4
	Sampling program	5
	Water sampling	9
RESUI	LTS	10
	Cross sections in 1st track	11
	Summary of 1st track	20
	Cross sections in 2nd track	22
	Summary of 2nd track	28
	Velocity of Strontium and Cesium through Soil	30
	Tracer tests	31
	pH tests Retention characteristics of soil	32 33
	Retention characteristics of soil	33
DISCU	SSION	34
CONC	LUSIONS	39
ACKN	CWLEDGEMENTS	40
REFE	RENCES	41
APPE	NDIX A	42
	Figures	
1	Water table beneath liquid disposal area	2
2	Water table at 'A' Disposal Area - 1950/1960	3
3 - 6	Multiple soil sampler	6
7	Plan of drilling program	8
8	Plan of 1st track of seepage	11
9-15	Cross sections in 1st track	12-18
16 17	Longitudinal section through 1st track Plan of 2nd track of seepage	21 22
18-21	Cross sections in 2nd track	23-26
22	Plan of radioactive migration from Disposal Area 'A'	29
23	Longitudinal section through 'front' of Sr-90	30
24	Graph: Sampling depth v counting accuracy	46



General Plan showing AECL Project, Disposal Area 'A' and Perch Lake.

INTRODUCTION

The original disposal ground for solid radioactive waste from the Chalk River Project is situated three quarters of a mile from the Project. Known as the 'A' Disposal Area, it lies in a 3-acre clearing in a wooded depression where radioactive waste was buried in trenches, excavated in the sand. This area was opened in 1946 and closed in 1955, but during the last three years of its operation three lots of radioactive liquid were poured into the soil.

Routine examination of foliage from the surrounding vegetation and of surface water from nearby swamps l have shown that some radionuclides have moved outside the disposal area. They have been transported down to the water table by leaching and percolation and have subsequently migrated with the ground water.

This paper describes the techniques now used at Chalk River to trace the rate and pattern of movement of radionuclides through the soil. It describes an investigation that was carried out with the following objectives:-

- 1. To find the radioactive nuclides in the soil and determine their origin.
- 2. To locate the 'fronts' of the fission products and estimate their velocities relative to that of ground water.
- 3. To estimate the total curies of fission products already outside the Disposal Area.
- 4. To estimate when these radionuclides will emerge above ground and contaminate surface water flowing via Perch Lake to the Ottawa River.

INFORMATION AVAILABLE

Between 1954-58 attempts were made to trace the migration of fission products by sinking a net-work of dry aluminum tubes in the ground and monitoring the soil in situ by lowering down a geiger counter. This method was satisfactory for measuring gamma radiation but was unsuitable for detecting low energy beta particles owing to the self-absorption of the soil and the shielding of the aluminum tube. The pattern of migration was consequently poorly defined because small quantities of cesium-137 could be detected while much larger concentrations of strontium-90 went unnoticed.

A series of well-points had also been installed round the 'A' Disposal Area and water samples from these had been regularly examined. Some had become contaminated with radiostrontium thus indicating the approximate direction of its passage from the disposals. Further evidence of the direction of seepage had been found from a hydrological survey covering South and Perch Lake swamps that lay south of Disposal Area 'A'. This had shown that the ground water flow beneath the solid disposal area was being affected by the proximity of the Liquid Disposal Area.

The routine disposal of large volumes of low level active water is made into pits that have been excavated in sand dunes and filled with aggregate; these are shown on Fig. 1 as the Liquid Disposal Area. In the period 1953-56, Reactor Pit 1 received 50-100,000 gallons/day and some of this seeped westward toward Disposal Area 'A' where it appeared in a spring before re-entering the soil via the disposal trenches. It was estimated that 2mC/day entered Disposal Area 'A' by this route. In 1956 Reactor Pit 2 was built and the flow was transferred from Reactor Pit 1 which was partially filled in and abandoned. Figure 1 shows how ground water has mounded beneath

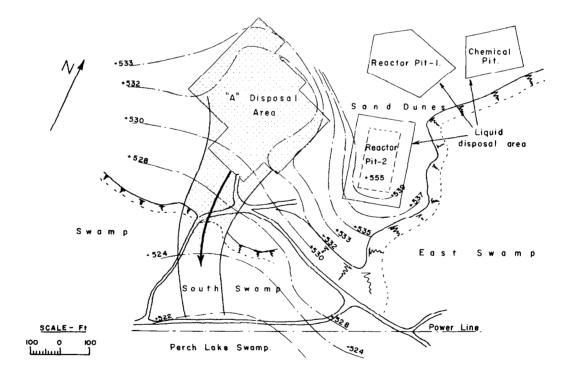


Fig. 1: Contours of water table shown distorted beneath 'A' Disposal Area by nearby Liquid Disposal Area Predicted direction and boundaries of seepage are indicated.

the new Reactor Pit altering the contours of the neighbouring water table beneath 'A' area. Since 1957 the percolating ground water from 'A' area has been diverted on its eastern flank and marshalled into a narrower course. The theoretical boundaries of the flow are shown in Fig. 1.

A ground water survey³ taken in 1950 beneath the original Disposal Area 'A' has been plotted in Fig. 2; the 1960 water table contours have been added for comparison. This shows that the ground water has generally risen over the period but that the hydraulic gradients have been reduced. The direction of seepage has probably changed greatest beneath the extended section of the disposal area.

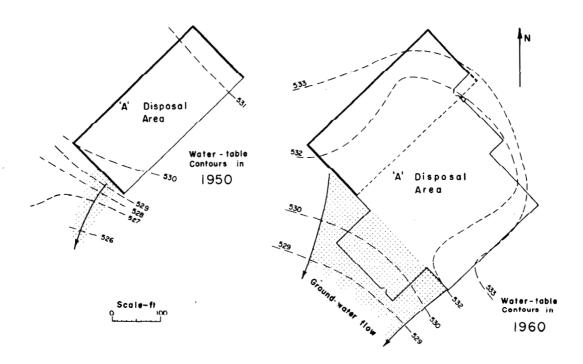


Fig. 2: Change in water table beneath 'A' Disposal Area between 1950 and 1960. (Area was enlarged in 1951)

It was expected that the principal migration would issue from the three liquid disposals in area 'A' all of which were deposited before 1957. Thus the search for tracks of radioactive migration would be spread over a wider band than that estimated from the present water table configuration (Fig. 1).

The first of the liquid disposals (1952) was an estimated million gallons of contaminated water that was fed into a trench system spanning most of the disposal area. The two other disposals were solutions of mixed fission products in water containing high concentrations of nitric acid and ammonium nitrate. Each was poured into a separate hole and may be considered as point sources of subsequent tracks of migration.

PROCEDURE

Previous information had shown that the main fission product to be located was strontium-90 and no method was known by which this radionuclide could be assayed in situ underground. Contaminated soil had to be examined in the laboratory for accurate assay and a detailed survey of the sub-soil was needed in order to collect the soil samples. The predicted path of seepage lay through South Swamp (Fig. 1) where the vegetation had become radioactive and formed part of a long-term series of observations. It was essential therefore that sampling operations or earthworks for access should not change the environment of the vegetation. Similarly, disturbance of the soil by driving operations had to be kept to a minimum. Experience with other sampling equipment had shown that the withdrawal of a 4-in. casing created an area of disturbed soil 6-8 ft in diameter and thereby affected its porosity. Finally, sampling equipment was required that would collect cohesionless soil (e.g. wet sand) from beneath the water table in samples of sufficient size for assay or radiochemical analysis.

Multiple Soil Sampler

A light portable tool known as the Multiple Soil Sampler was developed for this program. It is easily driven, simple in structure, and efficient in being able to collect several samples in one operation.

The sampler consists of a slender probe that is driven into the soil without a casing, (Fig. 3). It is assembled with connected lengths of hollow drill rods, (O.D. 1-5/16 in.), and as driving proceeds, additional sections may be added.

The sample receiving chambers are within the bore of the drill rods and are connected in series down the probe. Each chamber lies between an upper and a lower piston connected by a strut running axially up its centre, and soil may enter the chambers through slits cut at intervals in the side wall. When the probe is being driven, each slit (or port) is masked by the lower piston of each sample chamber, (Fig. 6). When the desired depth is attained the piston assemblies are hammered down to clear each port from the obstructing piston. The mobile soil particles flow into the sampling chamber with the ground water. The piston assemblies are then further depressed until the sample chambers are isolated from the surrounding soil by the upper pistons masking each port. The probe is then withdrawn and the soil samples collected as it is disassembled, (Fig. 5).

The sampler is driven into the soil using a portable automatic hammer powered by a 2-cycle gasoline engine, (Fig. 3). It is withdrawn using a crank operated jack and ball-cone clamp (Fig. 4) which, if required, can be used to press the sampler into the soil instead of using the hammer.

Over 2600 soil samples were taken throughout this investigation. Normally 12 samples were taken at one drive of the sampler but twenty have been collected on experimental runs. The maximum depth from which soil was sampled was 68 ft, this soil being the deepest from a string of eight samplers. The quality of the samples was compared with the gradings of undisturbed soil collected from adjacent positions with other equipment. This showed the silt content, normally about 10% of the soil, to be about 2% high owing to the vibration of the hammer.

Sampling Program

The normal procedure would have been to start sampling close to the disposal area and follow the tracks of fission products from their source. However, as it was winter when the preparations had been completed, sampling was started on the frozen swamp. This necessitated a broader initial search but allowed summer work to be carried out on higher and drier ground.

A 50-ft grid (300 ft long and 200 ft wide) was set out in South Swamp (Fig. 7) in the proper location to intercept the track anticipated from the hydrological survey². A boring was made at each intercept where eight samples were collected at 2-ft intervals between depths of 6 ft and 20 ft. Thirty seven borings were completed (290 soil samples) and radioactive soil was found in seven of these. Near the northern edge of the swamp individual soil samples showed slight activity but there was no



Fig. 3:
Driving sampler
with Atlas Cobra
hammer.



Fig. 4:
Extracting
sampler with
rack-jack and
ball-cone clamp.



Fig. 5: Collecting soil sample between double piston. (Drill rod previously removed.)

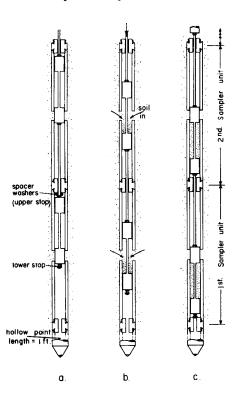


Fig. 6: Diagram of sampling method.

- (a) Pistons in position during driving.
- (b) Pistons depressed samples enter.
- (c) Pistons depressed further to close ports during withdrawal.

continuity between them. The seven boreholes showing major contamination were scattered diagonally across the grid and indicated that there was probably only one track of migration, (Fig. 7).

Borings were then driven 18 ft apart on a line estimated to be at right angles to the flow line. The radioactive soil samples were counted and the results were plotted to show a vertical cross section through the soil indicating depth and position of the highest concentration of radionuclides.

This procedure was repeated by sinking another line of borings parallel to the first, about 100 ft "downstream" and similarly finding the depth and spread of contamination. The line between these two centres was assumed to be the main track of seepage and a few borings were sunk along it to verify continuity between the two cross sections.

This line was then extended beyond the "downstream" section and borings made along it until the counts on the soil samples decreased to almost background level. In trying to locate the 'front' of radioactivity, borings were made as close as 1 ft apart along the line. Samples were taken on either side of it to ensure that the principal line of sampling lay on the longitudinal axis of flow. It was noted that the axis of migration through South Swamp lay within the predicted boundaries for seepage from the Disposal Area (Fig. 1). It was therefore concluded that the progression of fission products was following the theoretical curved path between their origin and the swamp.

Transverse lines were set out along this path at convenient intervals of 50 and 100 ft and borings were made along each of them. The sampling progressed northwards out of South Swamp, through the dune ridge and up to the perimeter fence of the Disposal Area. One line of boreholes was driven inside the fence (Fig. 7). A second course of radioactive seepage was intercepted near the disposal area; the transverse lines were extended and more borings taken in order to examine this track (Fig. 7, 2nd. track).

Borings were usually spaced 12-18 ft apart but if the depths of contamination were not similar in adjacent holes, extra soil samples were taken between them. In the long cross sections near the perimeter fence, borings were deep because of the height of the sand dunes and more numerous because of the complex distribution of contaminants.

A few samples were analyzed from each cross section to determine which fission products were present. When it was found that a radionuclide had traversed only certain sections, borings were made

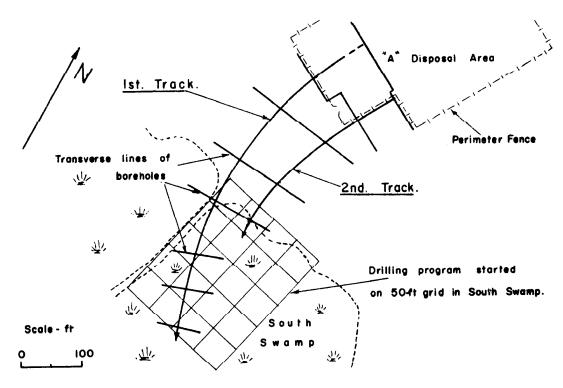


Figure 7:- Plan of drilling program showing grid and transverse lines of boreholes intercepting tracks of radioactive seepage.

along the flow-line 'downstream' from the last contaminated section, and the 'front' of its movement was detected by examining each jar of soil sample in a 100-channel gamma ray spectrometer.

Each soil sample was examined for texture and radioactive contamination. Only 0.050g was used for radioassay from an average dry sample weight of 60g; this was counted for total $\beta\gamma$ in a beta counter. Where more than one fission product accompanied Sr-90 a radiochemical analysis was carried out to determine the concentration of each radionuclide. However, if only one was present with Sr-90 and this was a gamma emitting isotope, the concentration of this radionuclide was determined by pulse height analysis. The combined counts from both radionuclides were measured in a beta counter and the quantity of radiostrontium deduced by difference.

Counting results from soil samples were plotted on cross sections

and the number of "nominal curies" of fission products present were calculated at each section. However, a total summation could not be completed until parallel estimates had been made for the fission products dissolved in the ground water. When the distribution of contamination on the soil was known, it was not difficult to select a few water-sampling points that would adequately reproduce the complementary cross section for ground water.

Water Sampling

Water samples were taken at 2-ft intervals corresponding to the levels where soil had previously been collected. They were collected by using modified porous bronze piezometers driven into the soil on a series of drill rods. Ground water could enter the body of the piezometer only through its porous bronze sidewall and was withdrawn by means of two small polyethylene tubes, one being attached to an evacuated bottle and the other left as a vent. This equipment had previously been used for sampling at one depth only⁴, and experiments were needed to adapt it to collect samples consecutively from various depths.

In the initial tests the piezometer was driven to the greatest depth required and water samples were collected every 2 ft during extraction. These samples were not satisfactory owing to contaminated water rising to the sampling point from lower positions previously vacated.

Further tests showed that if samples were withdrawn during driving instead of extraction, the hammer being stopped every two feet for this purpose, the only cross contamination came from water that was already in the point from a higher level. It was necessary, therefore, to increase the rate of withdrawal in order to drain this unwanted water from the point faster than the fresh sample could enter.

In order to drain the point rapidly (vol. = 100ml), experiments were conducted with larger diameters of plastic tubing. By increasing the diameter to 1/8 in., 96 ml could be lifted 25 ft in 29 secs and 4 ml remained as discontinuous bubbles in the tube. Larger diameter tubing required suction bottles greater than 500 ml and also left more water uncollected in the tube. The vent had to be the same diameter as the suction tube, because at the start of suction both tubes were filled with water up to the level of the water table.

Thin walled (0.020 in.) polyethylene tubing of 0.118 in. diameter was adopted. Withdrawal rates using a 500-ml suction bottle were satisfactory down to depths of 25 ft, but beyond that the suction had to be

supplemented by a slight positive pressure on the vent; this was supplied by nitrogen from a cylinder.

Results from the water samples were used to confirm the distribution of activity shown by the earlier data from soil samples. Complementary soil and water results were plotted on each vertical cross section and the contamination on soil and in water was calculated for the volume bounded by the section and having an assumed thickness of one foot. These results were further sub-divided to determine the curies of each radionuclide in this volume and were calculated in curies/foot. Since the cross sections were usually at right angles to the flow path, each result represented the gross concentration of radionuclide along a unit length of track. The total of fission products distributed along a track was computed from the concentrations at each cross section.

Rate of seepage through soil

One year after the radioactive 'front' was found under South Swamp, samples of soil were again taken in this area and the new position of the 'front' was found. By measuring the distance travelled in one year, seasonal variations in the flow-rate of ground water could be disregarded.

The velocity of ground water was measured at two positions along the longest (1st) track of active seepage. Fluorescein and sulphur-35 were injected into the ground water through porous bronze points and their rates of movement measured by sampling ground water to find the tracers from positions "downstream".

RESULTS

Details of the two tracks of radioactive seepage have been studied by reference to their cross sections. The concentration of radionuclides in each cross section have been calculated and the results summed to yield the total fission products in each track. A typical calculation for a cross section has been included in Appendix A.

A statistical test was made to estimate the probable accuracy of these results since only 0.050g of soil was counted from each 60g sample. These results are shown in Appendix A. In each of the cross sections to be examined, estimates of the curies of contamination have been given limiting values indicating the possible error in curies, based on the standard deviation of counts from the relevant samples.

Examination of Cross Sections

For convenience the longest track of seepage associated with the 1954 (nitrate) disposal will be called the 1st track and that from the 1955 (acid) disposal will be the 2nd track; they will be dealt with in this order.

Cross Sections Along First Track

In the 1954 disposal, 1500 gallons of active liquid were fed into a hole 10 ft deep in the sand. The mixed fission products totalled 173 curies and contained 60 curies of Sr-90 and 70 curies of C_8 -137 in solution with 3/4 ton nitric acid and 2 1/4 ton ammonium nitrate. Lime and limestone were placed in the bottom of the hole in an attempt to neutralize the acid solution. The positions of the eight cross sections (AA-HH) associated with the first track are shown in Fig. 8.

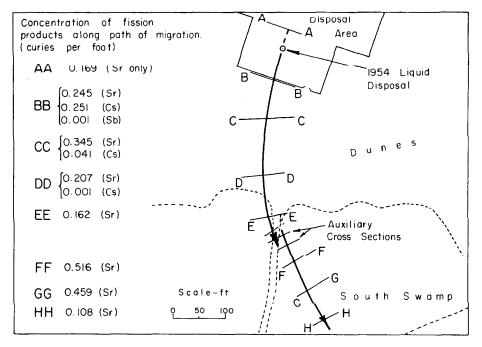


Fig. 8: Plan of cross sections on 1st track of seepage. Details of fission products at each cross section are also shown.

Cross Section AA (Fig. 8)

Cross section AA was sited 'upstream' from the 1954 liquid disposal to intercept the seepage from most of the remainder of the disposal area. Results showed that strontium had spread fairly uniformly through the sand beneath ground water level and that its total concentration passing through the section was 0.17 ± 0.034 curies/ft. There were no other contaminants present at this location.

Cross Section BB (At perimeter fence) (Fig. 9)

Cross section BB lay along the line of the perimeter fence 100 ft 'downstream' from AA and 60 ft 'downstream' from the nitrate disposal. Figure 9 shows that contamination began at 11 ft below ground level at the water table and penetrated 24 ft deeper through the sand. At this depth, silt deposits, being less permeable, prevented further vertical dispersion. The width of the path of strontium was about 95 ft and it contained 0.24 ± 0.050 curies/ft. A comparison of these results with those from section AA

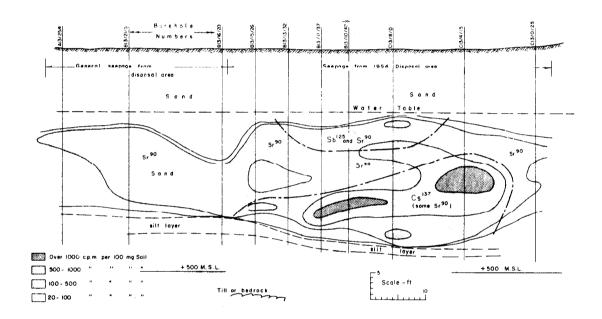


Fig. 9: Cross section BB. Beneath perimeter fence of Disposal Area. Sr-90 contaminates the whole of the shaded area of soil. Regions containing Cs-137 and Sb-125 are indicated.

shows that most of this had come from general seepage and probably only 25% was from the nitrate disposal.

Other contaminants of the soil were antimony-125 (0.001 C/ft), situated close to the water table, and cesium-137 (0.25 \pm 0.059 C/ft) deeper down. These originated from the nitrate disposal and showed that seepage had dispersed into a 60-ft wide band only 60 ft from its origin. Fission products in the water of this section totalled 0.0052 C/ft.

Section CC (70 ft from perimeter fence) (Fig. 10)

Here the fission products had moved downwards; soil was uncontaminated down to eight feet beneath the water table and the radioactive regions were contained over deposits of varved clay and silt. The band of Sr-90 had contracted from 1500 ft² to a cross sectional area of 1200 ft² and its concentration was higher at 0.34 \pm 0.072 C/ft. Cesium-137 was present at a reduced concentration (0.04 \pm 0.010 C/ft) but no antimony had reached this position. Water in this cross section contained 0.0044 C/ft of cesium plus strontium.

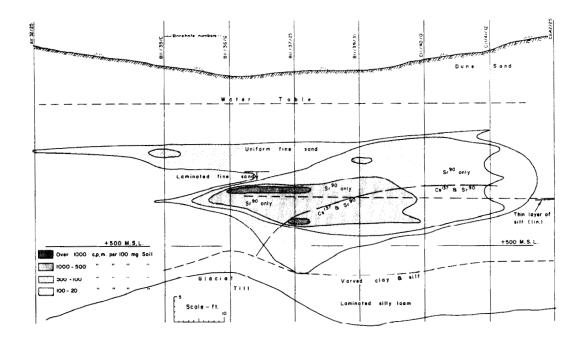


Fig. 10: Cross section CC. Where the track of radioactive soil passes beneath a hollow in the sand dunes.

Section DD (180 ft from perimeter fence) (Fig. 11)

Section DD was on the last line of borings through the sand dunes. Here, the active band had been further constricted to 600 ft² between the water table and a rising bed of varved silt. Its top remained eight feet beneath the water table and its width was unchanged. Only a small concentration of cesium was present (0.001 C/ft) and the concentration of strontium had decreased to $0.21 \pm 0.047 \text{ C/ft}$. The total content of Sr-90 in the water of this section was 0.0028 C/ft.

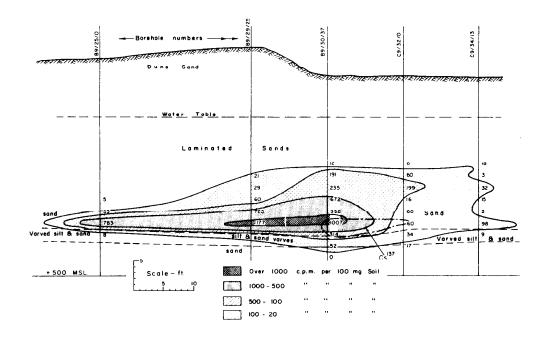


Fig. 11: Cross section DD, near the base of the sand dunes.

Radioactive water passes through sand over layers of varved silt.

Section EE (267 ft from perimeter fence) (Fig. 12)

Section EE lay beneath an access road dividing the west swamp from South Swamp. The pattern of concentration at this section was depressed beneath the road, the bulk being beneath the west swamp where its upper limit was only 3 ft below grade; however, the surface water was inactive. The flow area had expanded owing to a dip in the silt deposits and the rise of the flow net to swamp level. This section was beyond the 'front' of cesium movement and strontium was the only radionuclide present. Its gross concentration was 0.16 ± 0.015 C/ft, the lowest of the sections so far examined. The gross activity in the water was 0.0018 C/ft.

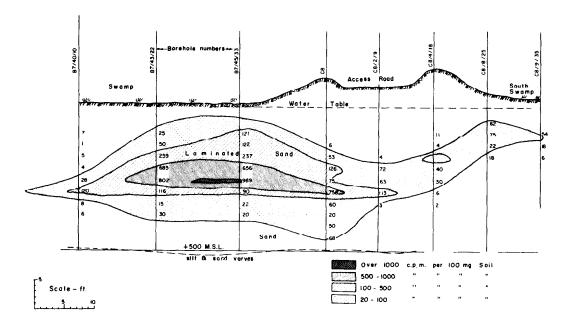


Fig. 12: Cross section EE under road between west and south swamps. Only Sr-90 has reached this position.

CRER-1018 - 16 -

Cross Section FF (367 ft from perimeter fence) (Fig. 13)

A comparison of Figs. 12 and 13 indicated that a major change in the distribution of contaminants had occurred somewhere between Sections EE and FF. In FF a broad mass of strontium was found beneath the surface of South Swamp and its strontium concentration of 0.51 ± 0.035 C/ft was nearly three times as high as in the previous section (EE). Because of the disparity between Sections EE and FF, additional lines of borings were sunk between them and auxiliary cross sections were drawn.

Results indicated that the thinning path of strontium that had passed through Sections AA to EE had penetrated the tail of a broad mass of strontium that lay at a slightly higher elevation beneath South Swamp.

The bulk of the strontium in Section FF lay 8 ft beneath the surface but part of this rose to the surface and was exposed in the humus in a band 30 ft wide. The humus has a higher affinity than sand for strontium so that the activity of this material as it appears in the cross section distorts the true shape of the dispersion by ground water.

The gross activity in the water of this section was 0.0093 C/ft.

Section GG (427 ft from perimeter fence) (Fig. 14)

Section GG intersected the same large mass of Sr-90 described in the previous section; its gross contamination was 0.46 ± 0.039 C/ft. The region of highest concentration was about 40 ft wide in a band between 9 and 15 ft below ground level. The gross activity in the water was 0.0080 C/ft.

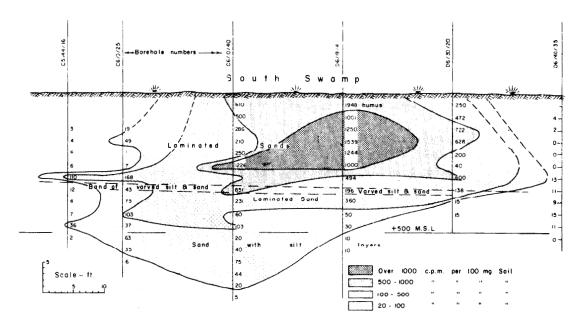


Fig. 13: Cross section FF in South Swamp taken through mass of Sr-90. High concentration in organic material near surface.

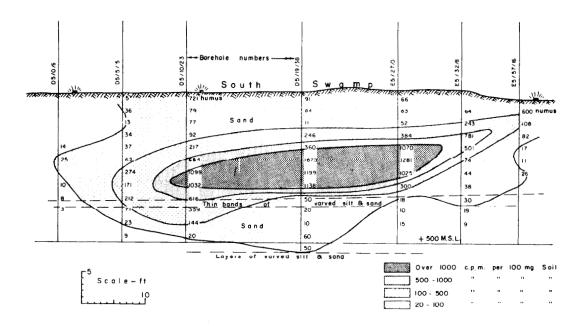


Fig. 14: Cross section GG in South Swamp through mass of Sr-90 concentrated between 9 and 15 ft below the surface.

Section HH (500 ft from perimeter fence) (Fig. 15)

The last section, HH, lay about 65 ft behind the front of strontium movement. There was no highly contaminated soil (>1000 c.p.m./100 mg) and the total strontium content was 0.10 ± 0.007 C/ft. The main body of strontium lay 11 ft beneath the swamp but some had extended to the organic material at the surface and was exposed in a 30-ft wide band.

The water of the section contained 0.0015 C/ft.

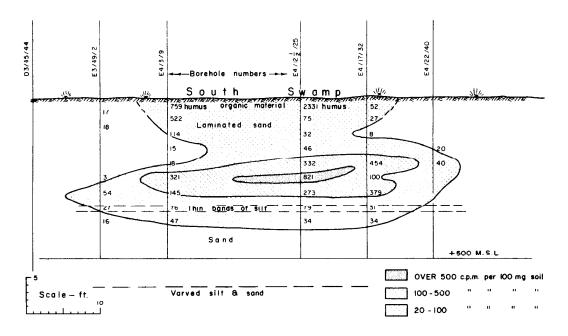


Fig. 15: Cross Section HH in South Swamp showing distribution of Sr-90 in soil 65 ft behind the "front".

Results from these eight sections were analyzed to determine the total quantities of each fission product outside the perimeter fence. Table I deals with quantities in the first track to South Swamp and Table II with the quantity of Sr-90 in South Swamp.

Table I - Calculation of fission products in 1st Track. (Between Disposal Area & South Swamp).

NO	ICE	mal)	Concentration of Activity		Activity between Sections		
CROSS	DISTANCE APART	SKEW (Off Normal)	at Section	Normal to flow	Mean Conc.	Gross	TOTAL Fission Product in (1954) track
		٠	(C)	C Cos θ			·
	ft	θ°	Curies per ft	Curies per ft	Curies per ft	Curies	
					STRONTIU	JM IN SO	IL
BB	75	0	0.245	0.245	0.2010	21.005	
CC	75	11	0.345	0.3388	0.2919	21.895	
DD	111	8	0.207	0.2049	0.2718	30.175	
EE	81	0	0.162	0.1620	0.1834	14.859	
	90				0.0810	7.290	74.216 Curies
					CESIUM	IN SOIL	
BB	75	0	0.251	0.251	0.1450	10.000	
CC	-	11	0.041	0.0402	0.1456	10.920	
DD	111	8	0.001	0.0099	0.0251	2.786	
Front	30		0	0	0.005	0.150	13,856 Curies
					ÄNTIMONY	IN SOIL	
BB		0	0.001	0.001	1		
Front	50		0	0	0.0005	0.025	0.025 <u>Curies</u>
					VATER	(Total $\beta_{\mathcal{Y}}$)	
BB	75	0	0.0052	0.0052	0.0048	0.357	
CC	111	11	0.0044	0.0043	0.0035	0.394	
DD	81	8	0.0028	0.0028	0.0023	0.185	
EE	90	0	0.0018	0.0018	0.0009	0.081	1.017 Curies
			0	0	0.0003	0.001	1.017 Outes

Table II - Calculation of fission product in bulk of contamination in South Swamp.

7	ш		centration Activity			
CROSS	DISTANCE	at Section	Mean between sections	Gross Activity between sections	TOTAL. Fission Products of mass (5r) in	
SS	A T.	Curies per ft	Curies per ft	Curies	South Swamp	
			SOIL	(all Sr-90)		
FF GG HH Front	70 50 68 7 0	0 0.523 0.459 0.108	0.2615 0.491 0.3335 0.054	18.305 29.460 22.678 3.780	74,323 Curies	
			WATER	(all Sr-90)		
FF GG HH Front	70 60 68 70	0 0.115 0.080 0.0015	0.00575 0.00975 0.00475 0.00075	0.4025 0.5860 0.3230 0.0525	l,364 Curies	

Summary for first track

Since seepage from much of the disposal area percolates past the site of the 1954 (nitrate) disposal, the resulting track of contamination contained strontium from both sources and it was not possible to distinguish the two fractions.

There were 150 curies Sr-90 in the first track. The portion that had migrated farthest lay as a mass of 75 curies beneath South Swamp (Table II). A 2-ft thick layer of humus at the surface contained 1.3 curies and the ground water held 1.4 curies in solution. The remainder was adsorbed on the soil about 10 ft down. The rearward portion lay along a continuous tongue of radioactive soil extending from the perimeter fence to South Swamp (Table I). This contained 75 curies Sr-90, 14 curies Cs-137 and 0.02 curies Sb-125.

In figure 16 two longitudinal sections have been drawn. The first indicates the distribution of each fission product and the second shows the strata of unconsolidated deposits. This information was derived from three

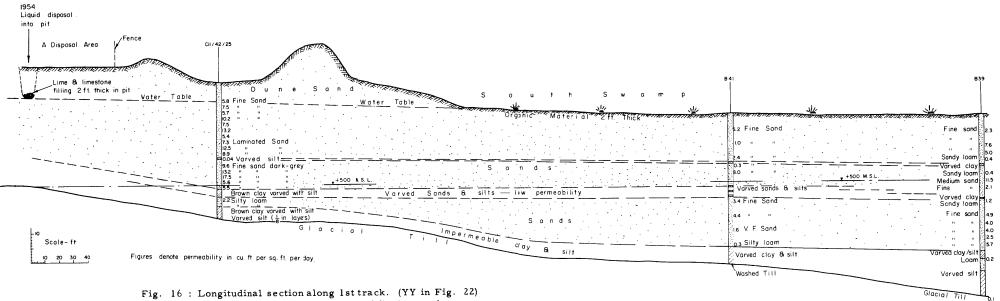


Fig. 16: Longitudinal section along 1st track. (YY in Fig. 22)

Upper section: Distribution of fission products.

Lower section: Distribution of soil deposits.

boreholes from which undisturbed soil samples were collected² and tested for permeability. All the fission products in the first track lie within the top band of sand, approximately 25 ft thick, that overlies strata of less permeable varved silts. It will be shown later that fission products from the 2nd track have penetrated the sands beneath this silt and that they are trapped there in a confined aquifer.

Cross sections along second track

The 2nd track issues from the 1955 (acid) disposal where 11,000 gallons of active liquid (2.2 x 10⁶ c.p.m./ml) were fed into a hole in the sand. The mixed fission products, approximately 900 curies, contained 300 curies Sr-90, 250 curies Cs-137 and 120 curies Ru-106. They were in solution with nearly 9 tons of nitric acid (2.5N) and no attempt was made to neutralize this. Borings were made on five transverse lines along this tongue of seepage.

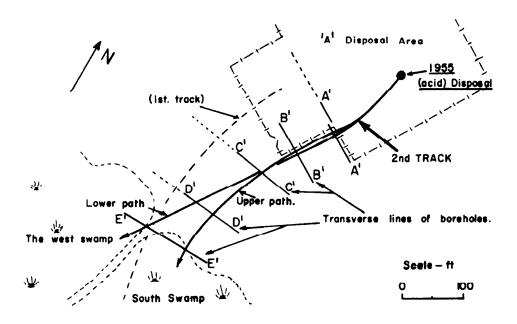


Fig. 17: Positions of cross sections along seepage path from 1955 disposal (2nd Track)

Cross section A'A' (at perimeter fence 140 ft from 1955 acid disposal)

Most of the contaminants in this section had originated from the 1955 (acid) disposal and the region of high concentration was about 50 ft wide. Between the disposal point and line A'A' lay a trench filled with solid radioactive waste; some of the contamination in section A'A' may have come from this source.

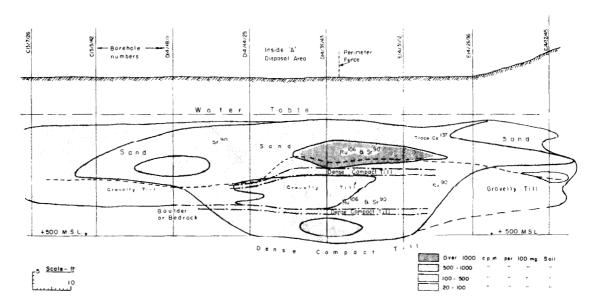


Fig. 18: Cross section A'A' 140 ft from 1955 (acid) disposal beneath perimeter fence has the highest concentration of Sr-90 of all the cross sections. (1.26 curies/ft)

Fission products first appeared 12 ft below ground level at the water table, and were present for a further 31 ft through sands and a lens of gravelly till (Fig. 18). Dense compact till had prevented further penetration and the band of contamination was 140 ft wide. Sampling was difficult within the lens where most drives were halted by large boulders.

The strontium concentration (1.26 \pm 0.17 curies/ft) was the highest of any section examined; this was roughly divided into two, with one half going over the lens and the remainder percolating through it. There was a low concentration of ruthenium-106 (0.004 \pm 0.0007 C/ft) and a trace of cesium-137. The gross activity in the water of the section was (0.004 C/ft).

Cross section B'B' (80 ft from perimeter fence) (Fig. 19)

The track of active soil had resolved into two layers. The upper layer, between 18 and 28 ft below grade, contained 0.24 ± 0.04 C/ft in a band 80 ft wide; a trace of cesium was present. The lower layer, between depths of 32 and 44 ft, contained 0.31 ± 0.10 C/ft Sr-90 and 0.005 ± 0.0017 C/ft Ru-106; this band was 96 ft wide. Radioactive ground water in this layer had gravitated to impervious silt deposits overlying glacial till, and spread laterally over it. The gross activity in the water of the section was 0.0041 C/ft.

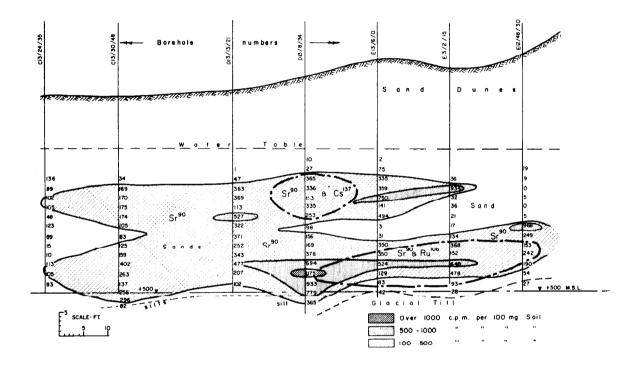


Fig. 19: Cross section B'B' showing a division of the radioactive seepage into two layers with the lower layer spreading laterally over glacial till and silt.

Cross section C'C' (146 ft from perimeter fence) (Fig. 20)

The seepage pattern had remained in two layers. The upper track, containing strontium only, had dwindled to 0.055 ± 0.014 C/ft and was 40 ft wide. The cross section was inclined 11° off normal to this axis of flow but was 25° off normal to the direction of seepage in the lower section.

The concentration of strontium in the lower track was unchanged $(0.320 \pm 0.126 \text{ C/ft})$ and a slight trace of ruthenium indicated that its 'front' lay close to this position. No cesium was present. The gross activity in the water of the section was 0.0025 C/ft.

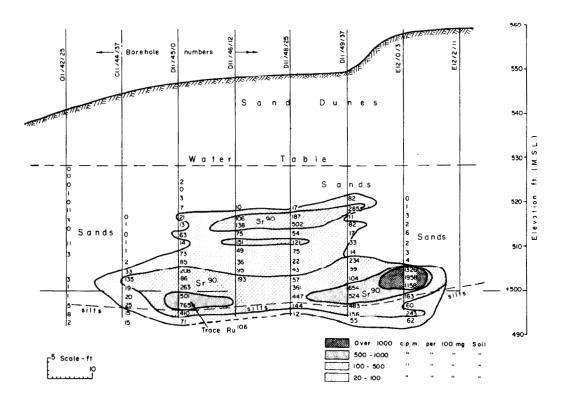


Fig. 20: Cross section C'C' showing two layers of concentration of Sr-90. The 'front' of Ruthenium-106 occurs near this point.

Cross section D'D' Upper track 251 ft from fence (Fig. 21) Lower track 266 ft from fence

The two layers had continued to diverge and at this section their flow axes were 32 ft apart. Strontium was the only fission product present and its gross concentration was 0.057 ± 0.015 C/ft in the upper band and 0.056 ± 0.017 C/ft in the lower band. The section was inclined 30° off normal to the lower track where contaminated water had penetrated sands lying beneath silt deposits. The gross activities in the water of the section were 0.0009 C/ft (upper track), and 0.0003 C/ft (lower track).

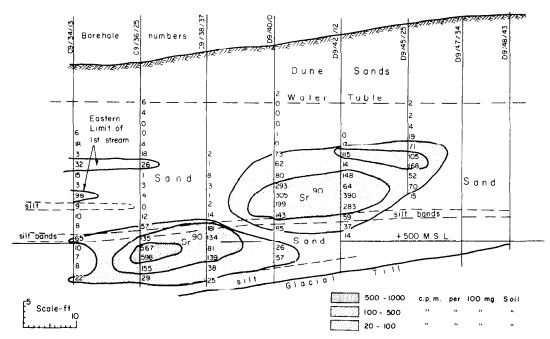


Fig. 21: Cross section D'D' showing two diverging layers. The lower layer is moving to left (out of paper) and is trapped beneath silt bands.

Section E'E'

The upper and lower tracks had diverged and their centres were 100 ft apart. The upper track was 326 ft from the perimeter fence and the section lay just within South Swamp. Radioactive soil was 16-23 ft beneath swamp level with a gross concentration of strontium of only 0.010 C/ft. Narrow tongues of low contamination (20 c.p.m./100 mg) had spread through the soil in a 30-ft wide band. At one end a small region of higher concentration was the root of a narrow tongue stretching 65 ft further to its 'front'. However, section E'E' represented the 'front' for the greater part of the upper track.

The lower track (366 ft from perimeter fence) had continued in a straight line. It was sufficiently deep to pass beneath the 1st track, previously described (Section EE), and had penetrated the west swamp. It

lay beneath silt bands at a depth of 34-36 ft with a concentration of 0.019 C/ft of Sr-90. The 'front' of this track was 70 ft beyond this cross section and 38 ft beneath the surface of the swamp.

Combined results from cross sections A'A' to E'E'

Table 3: Total Fission products in radioactive track (outside perimeter fence) from 1955 (acid) disposal.

z	й	a		Concentration Activity between of Activity Sections			
CROSS	DISTANCE	SKEW (Off Normal)	at Section	Normal to flow	Mean Conc.	Gross	TOTAL Fission products in track from 1955 (acid) disposal
	ft	θ°	(C) Curies per ft	C Cos θ Curies per ft	Curies per ft	Curies	outside perimeter fence
					S	r-90 in Soil	(Upper Track)
A'A' B'B' C'C' D'D' E'E' Front	80 66 105 75 65	0 0 11 0	0.645 0.245 0.055 0.057 0.010	0.645 0.245 0.054 0.057 0.010	0.4450 0.1495 0.0555 0.0335 0.0050	35.600 9.867 5.827 2.512 0.325	54.131 Curtes
A'A' B'B' C'C' D'D' E'E' Front	80 66 120 100 70	0 0 25 30 0	0.617 0.315 0.320 0.056 0.019	0.617 0.315 0.288 0.048 0.019	0.466 0.3015 0.1680 0.0335 0.0095	37.280 19.899 20.160 3.350 0.655	(Lower Track)
A'A' B'B' C'C'	66 80	0	0.0038 0.0052 0	- -	0.0045 0.0026	0.360 0.172	UM IN SOIL U.532 Curies
A'A' B'B' C'C' D'D' E'E'	80 66 105 (80)	0 11 0	0.0043 0.0041 0.0025 0.0012 0.0001	0.0043 0.0041 0.0024 0.0012 0.0001	0.0042 0.00325 0.0018 0.00065	0.336 0.214 0.189 0.052	TOTAL βγ)

Summary for 2nd track

Results from sections A'A' to E'E' have been combined in Table 3 and the quantity of each radionuclide in the track was calculated. This track issuing from the 1955 (acid) disposal contained 135 curies of Strontium-90 outside the perimeter fence of 'A' disposal area. Half a curie of Ruthenium-106 had moved along the track and its 'front' was about 150 ft outside the fence. Trace quantities of cesium-137 were also found.

The track divided into two; the upper layer containing 54 curies of Sr-90 lay along the predicted curved path and extended 390 ft from the fence. Its 'front' lay 19 ft beneath the surface of South Swamp. The lower layer of strontium, initially in a solution of higher specific gravity, had spread out in a thin layer over less permeable till. As the till dipped towards South Swamp the liquid followed it down and entered sands lying beneath a band of impermeable silt. It had travelled in a straight line in this confined aquifer. The front was nearly 440 ft from the perimeter fence and was 38 ft beneath the surface of the west swamp. Eighty-one curies of Sr-90 were distributed along this track.

The combined results of studies on both tracks are plotted in Fig. 22. Section lines and concentration data have been omitted for clarity. Heavily shaded areas show the location of high concentrations of radionuclides and are not strictly quantitative but the outlines of shaded areas are a precise delineation of the migration pattern observed early in 1961.

Fig. 22: Plan of radioactive seepage of ground water from 'A'
Disposal Area. YY is line of longitudinal section in Fig. 16

29

Velocity of Strontium and Cesium through Soil

In 1960, soil samples were taken along the flow axis of the principal mass of strontium (1st track) in South Swamp. Borings were made behind and ahead of the 'front' and contours of strontium concentration were drawn. It was found that the anticipated accuracy in pin-pointing the 'front' (± 1 ft) could not be achieved owing to a series of sinuous narrow tongues of low-level contamination that preceded the main body of strontium.

One year later soil samples were again taken from some of the original borings and new contours of strontium concentration were established. (see figure 23) The 'fronts' of strontium at 1000 c.p.m. and 100 c.p.m./100 mg had advanced 15 ft in one year.

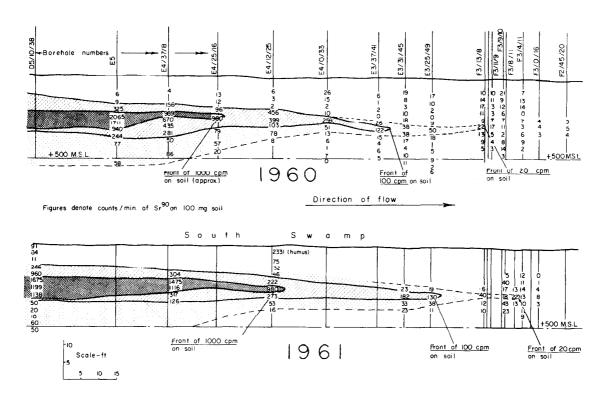


Fig. 23: Longitudinal cross sections through 'fronts' of strontium.

Shaded regions (>1000 and >100 c.p.m./100 mg) have
advanced 15 ft in one year.

The front of cesium was found 216 ft from the perimeter fence in July 1960. (see Figure 22) In May 1961 more soil sampling showed that it had advanced about 6 ft. Its rate of migration is probably between 7 and 7.5 ft per year.

Tracer Tests

The velocity of ground water near the strontium 'front' was measured using sulfur-35 and fluorescein as tracers. The tracer solution was injected through a series of porous bronze piezometers sunk to the same depth as the strontium but in neighbouring uncontaminated soil. The tracer solution lay in a band 3 ft wide x 1 ft deep and 100 ml samples were withdrawn from positions "downstream" by the water sampling methods already described.

Results showed that the ground water travelled at 1.54 ft/day measured over a distance of 20 ft. A measure of the delay of strontium by adsorption on the soil is given by the ratio.

$$\frac{\text{Rate of ground water flow}}{\text{Rate of Sr-90 advance}} = \frac{1.54 \text{ X } 365}{15} = 37.5$$

or Rate of Sr-90 movement = 0.027 X vel. of ground water

A similar test was conducted in the centre of the 1st track 70 ft from the perimeter fence (on section CC). Because the ground water was heavily contaminated with strontium and cesium, the radioactive tracer was omitted and fluorescein only was used. A total of 32 litres of water was injected through 6 points at 8-in. centres into a band 4 ft wide x 2 ft thick. The injection concentration of dye was 0.5 g per litre. The velocity of the dye was 1.67 feet per day taken over 15 ft in three 5-ft measurements.

Other experiments 6 have shown that the velocity of fluorescein is 0.75 x velocity of ground water in this soil. Thus the effective velocity of the ground water in the 1st track near the perimeter fence was $\frac{1.67}{0.75} = 2.23$ ft/day

Comparison with water table contours

The ratio of the velocity of ground water at the perimeter fence to that in South Swamp was

$$\frac{V \text{ fence}}{V \text{ swamp}} = \frac{2.23}{1.54} = 1.45$$

This result is in agreement with the ratio of water table gradients (1/75 & 1/120) at these points, giving $\frac{120}{75}$ = 1.6

A fluorescein test (2) on section AA inside the disposal area and 40 ft upstream from the nitrate disposal has shown that the velocity of the ground water movement there was 0.50 ft/day. Another test was carried out on the predicted path of seepage through Perch Lake Swamp. This was 1300 ft ahead of the present 'front' of strontium. Fluorescein was injected through 6 points to the expected depth of migration at 8 ft and the velocity of the dye was found to be 0.75 - 1.0 ft/day.

Tracer experiment using dye, Sr-90 and ammonium nitrate

A field experiment was carried out in a neighbouring inactive area in which radioactive water, withdrawn from the strontium mass in South Swamp, was mixed with fluorescein and ammonium nitrate and injected 12 ft into the soil. Results showed that the presence of ammonium nitrate accelerated the speed of migration of strontium to half that of fluorescein, i.e. by a factor of nearly 20. Thus $\frac{V \text{ water}}{V \text{ strontium}} = \frac{1}{0.5 \times 0.75} = 2.66$

Experiments by Merritt⁷ at the same point had shown that

V water V etroptium * 50 in normal ground water without added ionic material.

pH Tests

Ground water was sampled from points in the 1st and 2nd tracks and in the mass of Sr-90 in South Swamp and tested for pH with the following results:

(1)	1st track	60 ft from	1954 nitrate dispo	sal $pH = 6.0$
-----	-----------	------------	--------------------	----------------

(2) 2nd track 140 ft from 1955 acid disposal
$$pH = 6.4$$

(3) From mass of
$$Sr-90$$
 in South Swamp $pH = 6.7$

This showed that the ground water was slightly acid in the radioactive tracks, but no different from normal ground water in the area, which has a pH between 5.4 and 6.6.

Retention characteristics of soil in South Swamp

In cross sections EE to HH, where strontium was the only fission product present, figures have been obtained of total quantities of Sr-90 adsorbed on the soil and remaining in solution. These results are based on a soil porosity of 38% with a specific gravity of the grains of 2.67.

The distribution ratio⁸ D =
$$\frac{\text{Total Sr-90 on soil of cross section}}{\text{Total Sr-90 in water of cross section}}$$

= $\frac{\text{c.p.m./g soil x 0.62 x 2.67}}{\text{c.p.m./ml water x 0.38 x 1}}$

but the equilibrium distribution coefficient $K_d = \frac{\text{c.p.m./g Soil}}{\text{c.p.m./ml Associated Water}}$

Thus
$$D = \frac{0.62 \times 2.67}{0.38} \text{ K}_d = 4.35 \text{ K}_d$$

Kd values for strontium from cross sections of 1st track in South Swamp

Table 4 : Calculated Values for K_d (Sr-90)

Cross Section	Curies Sr-90 along 1 ft of seepage path		$D = \frac{Sr-90 \text{ soil}}{Sr-90 \text{ water}}$	$K_{d} = \frac{D}{4.35}$	
	Soil	Water			Remarks
DD	0.2075	0.0028	73.0	16.8	Part of continuous
EE	0.1620	0.0018	90.0	20.7	tongue down to swamp
FF	0.5139	0.0093	55. 2	12.7	Mass of Sr-90 in
GG	0.4570	0.0080	57.1	13.2	South Swamp
НН	0.1000	0.0015	66. 1	15.2	Swamp

These results show that values for K_d (Sr) in the swamp mass are smaller than in the remainder of the track, despite similar soil properties. Also the K_d values in South Swamp vary inversely with the strontium concentration.

Kd (Sr) values from velocity of 'front'

Another estimate of the K_d (Sr) may be made from the relative velocities of ground water and Sr-90 at the 'front'. Using the following equation 9 for estimating the rate of advance of the adsorption 'front'.

$$Kd \rho + \epsilon = \frac{V \text{ water}}{V \text{ Sr}}$$

where ρ = dry bulk density (i.e. g dry grains/cm³ mixture) and ϵ = $\frac{Pore\ volume}{Total\ volume}$

..
$$K_d$$
 (0.62 x 2.67) + 0.38 = 37.5

$$K_d = \frac{37.1}{0.62 \times 2.67} = 22.4$$

This value for K_d is similar to that from section EE near the 'front' of the continuous tongue. (Table 4)

$K_{\mbox{\scriptsize d}}$ values for Sr-90 in cross sections of the 2nd track

Table 5: Calculated Values for Kd (Sr-90)

Cross Section		r-90 along page path	$D = \frac{Sr - 90 \text{ soil}}{Sr - 90 \text{ water}}$	Кd	
	Soil	Water			
B'B'	0.5600	0.0041	137	31.5	
Upper* C'C'	0.0546	0.0005	109	25.1	
Lower C'C'	0.3198	0.0020	160	36.8	
Upper D'D'	0.0572	0.0009	63.5	14.6	
Lower D'D'	0.0560	0.0003	187	43.0	

^{*}Upper & lower refer to the two layers of migration in this track.

- 35 - CRER-1018

These results show that the values for K_d (strontium) are much greater than those for strontium in the first track to South Swamp (Table 4). This is confirmed by the magnitude of the fraction of strontium that had migrated. Of 300 curies (Sr-90) in this disposal about 136 were outside the perimeter fence (i.e. 45%), whereas all the Sr-90 from the nitrate disposal was found in either the 1st track or South Swamp.

DISCUSSION

Cesium movement

It has been confirmed that the track of cesium originated from the nitrate disposal and that in 5-1/2 years it had moved 276 ft but in the next year it travelled only a further 7 ft. Measurements of ground water velocities and examination of water table contours have shown that the ground water velocity varies little along the total path (1.0 - 1.67 ft/day). It is therefore assumed that the cesium travelled swiftly following its disposal and that it has slowed up and probably reached an equilibrium velocity of 0.011 x velocity of ground water. It is known that nitrates travel rapidly with ground water and that the retention characteristics of the soil are sensitive to changes in pH and ionic concentration of the ground water. It is apparent that in its initial environment of calcium, ammonium and nitrate ions, the retention of cesium by the soil was low and that, following dispersion and dilution by ground water, the K_d has now increased to an equilibrium value.

Of the 70 curies of cesium in the disposal, only 14 had migrated beyond the fence (60 ft from origin) and 80% of this was within 75 ft of the fence. This indicated that only 4% of the cesium disposal seeped away rapidly after it was buried. The concentration of cesium varies inversely with its distance from the origin; this evidence supports laboratory tests showing that cesium becomes permanently fixed on the soil. This is probably because illite, a two-layer silicate clay mineral, is present in the clay fraction (<0.1%) of the soil⁴.

It was not practicable to obtain accurate field values for the K_d of cesium since this fission product has always occurred in regions heavily contaminated with strontium. However, spot checks on the relative concentrations of cesium in water and soil have shown that the K_d for cesium is approximately 100 in the 1st track.

Strontium Movement

Three ranges for the values of K_d (Sr) have been found. The lowest (12.7 to 15.2) occurs in the strontium mass in South Swamp. Since the highest concentrations of strontium distribution occurred in these cross sections (FF & GG), these figures agree with laboratory results showing that the value of K_d varies inversely with the concentrations of strontium. Values in the intermediate range ($K_d = 16.8 - 25.1$) have been calculated from medium concentrations of strontium in the 1st track, the upper layer of the 2nd track and from the velocity of the strontium 'front'; they are considered to be the K_d (Sr) values for the normal ground water of low ionic concentration. The high range of K_d (31.5 - 43) in the lower track from the acid disposal is an anomalous result, since laboratory tests have shown that an acid environment tends to decrease the value of K_d .

The track of general seepage from the disposal area passes the site of the 1954 (nitrate) disposal. Strontium sampled from "downstream" may have come either from this seepage or from the nitrate disposal; there is no way of differentiating between them.

The front of Sr-90 was 560 ft from the site of the 1954 disposal and in 1960 it advanced only 15 ft. It is probable that the conditions described for the early movement of cesium may also be applied to the strontium of that disposal. The present (1960) rate of migration at the front (0.027 x vel. ground water) is considered its equilibrium velocity in a normal environment, and is in agreement with results obtained in a field experiment using strontium-85.

It is suggested that the mass of strontium beneath South Swamp (74 ± 6 curies) originated from the 1954 (nitrate) disposal. Since this quantity represents rather more than the total estimate for that disposal, the remaining 74 curies (± 15) along the first track down to South Swamp, probably came from general seepage and may well be part of the 1000 curie disposal of water in 1952.

Strontium movement from 1952 (NRX) disposal

No evidence has been found of migration from this disposal of contaminated water, except in the track mentioned above.

In 1952, an estimated 1 million gallons of active water were poured into trenches (see figure 22); it was thought that the highest concentration was discharged at the beginning into a trench near the northern corner of the disposal area. When this started to fill, despite seepage, other interconnecting trenches were dug to accommodate the input, which lasted a month.

If the water with the highest contamination was discharged first, then its position in the north corner gave it the longest possible seepage path to South Swamp. The 2nd fluorescein test has shown that the ground water seepage is 6 in. per day on section AA; it would be less in the position of the trenches, where the hydraulic gradient is smaller. The contaminated water had a low ionic concentration so that a high retention of the strontium would have probably been realised. Assuming, for example, that the velocity ratio $(\frac{\text{water}}{\text{Sr-90}})$ was 37.5 (its present value at the 'front'), then in 9 years it will have travelled $9 \times 365 \times (\frac{0.5}{37.5}) = 44$ ft a value sufficiently low to show that most of this disposal could still be inside the perimeter fence.

It is most difficult to undertake a large sub-soil investigation inside the disposal area, owing to the high radiation fields, the accumulation of contaminated equipment on the surface and the maze of poorly charted burials.

Other Fission Products

Ruthenium 106 had previously been found in the surface water, in anionic form, where it had percolated from the liquid disposal area. In this survey it was found only on the soil in a cationic or neutral form. It is therfore assumed that this represented only a fraction of the ruthenium in the 1955 disposal and that the anionic fraction migrated rapidly soon after disposal. Antimony-125 is produced only as a small percentage of the total yield of fission products and because of this its discovery was a surprise. The 'front' was located too late to include measurements of its rate of migration in this report. It was expected that cerium-144 would be present among the migrating fission products, but it was not found.

Potential hazard from Sr-90 in South Swamp

The 74 curies of Sr-90 found under South Swamp is the largest potential hazard from these disposals, because it lies closest to free-running surface water. The mass of contaminated soil was 240 ft long and 65 ft wide and the regions of high concentration were between 9 and 16 ft below the surface. Humus in the top 2 ft of the swamp has a high affinity ($K_d \approx 400$) for strontium, which it has concentrated from comparatively low level active water near the upper boundary of the mass. The main mass of strontium is advancing through South Swamp and the 'front' will soon enter Perch Lake Swamp where it will be 1900 ft from Perch Lake.

A soil survey of Perch Lake Swamp² had shown that beneath its shallow surface of organic material, it is composed of sands that are laminated horizontally. These deposits are about 35 ft thick and overlie an impermeable stratum of clay; the water table is close to the surface. Because this unconfined aquifer has a regular cross section the flow lines will be horizontal and will rise to the surface only in the vicinity of Perch Lake. The previous estimate of the ground water velocity through Perch Lake Swamp² was calculated without direct measurements in the field. A new appreciation can be made using the measured rate of seepage 1300 ft downstream on the predicted path and at the anticipated depth (8-10 ft) beneath swamp level.

The water table gradient in Perch Lake Swamp (normally between 1/120 and 1/200) flattens out in the last 600 ft to the lake (1/380) and it is thought that a rise to the surface will occur in this region, which is often flooded Assuming that the rise will occur in the middle of this area (300 ft from lake) the Sr-90 is expected to remain submerged for another 1600 ft beyond its present position. An estimate in Table 6 has been made of the time taken for ground water to traverse this path.

Table 6: Travel time for ground water to traverse Perch Lake Swar

Length of path	Hydraulic Gradient	Ground Water Velocity	$\frac{\mathbf{v}}{\mathbf{i}}$	Travel Time	Total Time
ft	i	v ft/day		days	
100	1 120	1.54*	185	65	
490	$\frac{1}{163}$	(1.15)	(187)	427	
760	1 190	1.00*	190	760	
250	-	1.00(say)		250	1502=4.1 years

^{*}Measured rates of seepage.

The estimated time taken for ground water to travel through Perch Lake Swamp is about 4 years. Therefore the time taken for Sr-90 to travel from South Swamp to the surface near Perch Lake, assuming that it continues to move at $\frac{1}{37.5}$ x velocity of ground water, will be 37.5 x 4 or 150 years.

- 39 - CRER-1018

SUMMARY AND CONCLUSIONS

Although solid waste was buried in the 'A' disposal area for seven years before 1953, there is no appreciable radioactive leakage from this source. The principal radioactive contamination of the ground water has been caused by disposals of high-ionic liquid.

The first liquid disposal of the water from the 1952 NRX accident contained 1000 curies of Sr-90. A track of radioactive soil running out from the 'A' disposal area contains 74 curies Sr-90 which may have come from this disposal. However, most of the radionuclides remain inside the fence and probably not far from their disposal sites.

The experimental disposal of 1500 gallons of radioactive liquid containing 4600 lb ammonium nitrate, made in 1954, has resulted in the greatest movement of radionuclides under the ground.

The high concentrations of ammonium and nitrate ions caused all the radiostrontium to move off rapidly with the ground water to South Swamp but only 4% of the cesium was moved in this way. The movement was sufficiently fast to reduce vertical dispersion of the denser liquid and strontium remained close to the water table. Part of this rose to the surface when it entered South Swamp, but most of it remains submerged.

The mass of strontium is moving towards Perch Lake and will rise to the surface as it approaches or reaches the lake. Strontium-90 travels at 0.027 x velocity of ground water in South Swamp and continuing at this rate will take about 150 years before it rises to swamp level and disperses slowly into surface water. The quantity of Sr-90 will at that time have decayed from 74 curies through five half-lives to about two curies.

The fission products from the 1955 experimental acid disposal do not present a hazard. Of the principal fission products in this liquid (250C, Cs-137; 300C, Sr-90) no cesium and only 40% of the strontium have moved away from Disposal Area 'A'. The high specific gravity and retention by the soil have combined to produce an efficient disposal. The advanced 'front' of this migration less deep below South Swamp where it will cause no harm.

There is no danger of a mass release of fission products from 'A' area into surface waters. The site of the disposal area was well chosen; it lies close to the boundary of a ground-water basin and the ground water flow is slower than in neighbouring areas. The swamps of Lower Perch Lake Basin have good delay properties for Sr-90 and Cs-137 provided that their disposal is not accompanied by high ionic concentrations of liquid and that they are placed deep enough below the surface.

The exchange properties of strontium with this soil cause the migration of strontium to be delayed in the Perch Lake Basin for several half-lives. It is thought that the deeper deposits under-lying Perch Lake Swamp could be used as an underground reservoir for radioactive water.

ACKNOWLEDGEMENTS

The author wishes to thank Mr. W.F. Merritt for advice and assistance in the use of radioactive tracer techniques. Special thanks are offered to Miss E. Rawlence for her laboratory work and statistical studies and to Mr. G.G. Gunter for his field work.

REFERENCES

- 1. Ophel, I.L., and Fraser, C.D., 1957. The Chalk River Liquid Disposal Area, 1956, CRHP 709.
- 2. Parsons, P.J., 1960. Movement of Radioactive Wastes through soil. 1. Soil and ground water investigations in Perch Lake Basin. AECL No. 1038.
- 3. Pearce, J.A., 1950. Report on escape of activity from the disposal area. HRI-4.
- 4. Evans, E. J., 1958. Chemical investigations of the movement of fission products in soil. AECL No. 667.
- 5. Merritt, W.F., and Parsons P.J., 1960. Disposal of radioactive wastes. IAEA Vienna. Vol. 2 p 331-337
- Merritt, W.F., 1961. Movement of radioactive wastes through soil.
 Measurement of direction and effective velocity of ground water movement. AECL No. 1161.
- 7. Merritt, W.F., Private communication.
- 8. Newton, T.D., 1959. On the dispersion of fission products by ground water. AECL No. 909
- 9. Prout, W.E., 1959. Adsorption of fission products by Savannah River Plant Soil. U.S. AEC. DP-394.

APPENDIX A

Typical calculation for gross activity in a cross section

Samples within each borehole were collected at intervals of 2 ft. It was assumed that the counting result on a sample (from a known depth) was representative of the activity in the soil one foot above and below that horizon. By assuming that this level of contamination was maintained in the enclosing column of soil 1 ft sq. x 2 ft high, a summation of the column results for the complete borehole (2Σ c.p.m.) yielded a measure of its gross activity. By combining the mean of these borehole summations the gross activity was calculated for the complete cross-section with an assumed thickness of 1 ft. A final conversion was required to change the working units [c.p.m./100 mg)x ft³] to curies per ft run.

Table 7 shows the gross activities on soil and water in the soil columns surrounding each borehole, and Table 8 shows the summation of these results over the cross section.

It was noted that certain organic samples (humus) near the surface showed a very high counting rate dissimilar to that on neighbouring soil samples. A separate calculation has been made for this material.

Where no water samples were taken from a borehole the gross activity in the water was interpolated from the ratio total soil counts total water counts in the neighbouring boreholes.

Table 7: Typical summation of sample counts for gross activity on soil and water in boreholes of a cross section.

Elevation – ft M.S.L.	9/0/5G 10 Acti	vity on	soil s	hole	18'	bers 5	15'	tivity :	on wate	,	9, - 9, - 9, - 9, - 9, -	E5/37/16
Elevo	Soil	Soil	Water	Soil	Water	Soil	Water	Soil	Water	Soil	Water	Soil
523		5 [2	Humus 721	25	91	115	66	28			
521	i	36	18	79	256	84	133	63	; 31	64	154	Humus
519		13	2	77	133	11	36	52	10	243	Į.	108 -
517		34	16	92	71	246	261	384	622	781	728	82 -
515	14 -	37	27	217	12/	960	990	1070	715	501	412	17] —
513	25	62 1	115	654	,209	1675	1347	1281	919	74	15	11 -
511	17 —	274	364	1099	535	1199	1758	1025	379	44	23	26
509	10 -	171	373	1032	659	1136	71	300	24	38	16	1
507	81-	212	263	616	323	50	12	18	8	30	9	ţ
505	3 -	71	48	359	204	20	9	10	. 3	19	8	
503		23	29	144	. 55	10	5	15	4		ļ	
501	İ	9	1	20	10	60	30		:		}	
499		}				50	24					
ΣSoil ΣHumu	77	947		4389)	5592		4284		1794	4	244 500
ΣWater			1249		2582		4791		2743		1789	- 000
ΣSoil ΣWater	l (say)	0.76	6	1	.70	1	.17	1.	.56	0	.95	l(say)
2 ΣSoil	154	189			778		184	85			88	488
2 ΣWater 2 ΣHumu		249			164 442	95	82	54	36		78	488 1200
Z Zi iuiiiu	s –			1 '	144		_		-		-	1200

The last three lines in the table are measures of the total activity contained in a 1 ft sq column surrounding each borehole.

Table 8: Summation of borehole results (from previous table) for gross activity in typical cross section.

	Dist.		30IL.		WATER			нимиѕ		
Borehole number	'd' ft	2Σ ft³ >	Mean M	M x d /100mg	2Σ ft ³	Mean M × c.p.m.	M x d	2Σ ft³×	Mean M c.p.m.,	M x d /100mg
D5/0/6 D5/5/15 D5/10/23 D5/19/38 E5/22/0	10 10 18 15	154 1894 8778 11184 8563	1024 5336 9981 9876	10240 53360 179658 148140	154 2498 5164 9582 5486	1326 3831 7373 7534 4532	13260 38310 132714 113010 45320	0 0 1442 0	721 721	7210 12978
E5/32/8 E5/37/16	10	3588 488	6078 2038	60780 18342	3578 488	2033	18297	0 1200	600	5400
				470520			360911			25588

These results in $ft^3 \times c.p.m./100$ mg are converted to curies per ft. (i.e. the gross activity of the cross section assumed 1 ft thick.)

Conversion of c.p.m./100 mg soil into curies/sq ft cross section 1 ft thick.

Porosity of soil = 38% S.G. of soil = 2.67

Weight of soil grains/ft³ = 0.62 (12 x 2.54)³ x 2.67

=
$$\frac{4.6877 \times 10^4 \text{g}}{4.688 \times 10^5 \text{ c.p.m./ft}^3}$$

= 4.688 x 4.6 x 10⁵ d.p.m./ft³ (η * = 21.8%)

= $\frac{9.713 \times 10^{-7}}{4.688 \times 10^{-7}}$ curie/ft³

Similar calculations have been made for water and humus:-

Material	Porosity	S.G.	Conversion to Curies/ft3
Water with soil (above)		1	1 c.p.m./ml=2.229 x 10 ⁻⁸
Humus	90.2	1.62	1 c.p.m./100 mg=9.315 \times 10 ⁻⁸
Water in humus		1	1 c.p.m./ml=5.292 x 10-8

^{*}Counter efficiency for Sr-90 + Y-90 was the same as for Cs-137 + Ba-137.

Results for typical cross section.

Applying these conversion factors to the results from Table 8.

1. Gross activity in soil (all Sr-90)

=
$$470,520 \times 9.713 \times 10^{-7}$$
 = 0.457 curie/ft

2. Gross activity in water

=
$$360,911 \times 2.229 \times 10^{-8}$$
 = 0.0080 curie/ft

3. Activity in humus

=
$$25,588 \times 9.315 \times 10^{-8}$$
 = 0.0023 curie/ft

4. Activity in water in humus - negligible.

Statistical Tests

The concentration of gross activity on each soil sample was estimated from the counting rate of 0.050 g soil selected from a 60 g sample and a statistical test was made to estimate the probable accuracy of such results. Thirteen sample bottles, whose soil varied widely in contamination were examined. Twelve samples (0.050 g) were taken from each bottle and the standard deviation (per bottle) was calculated and expressed as a percentage of the mean count. (The coefficient of variation)

It was expected that the coefficient of variation would increase with the magnitude of the mean counting rate but this relationship was not established. Instead however, a correlation was noted showing that the coefficient of variation of the mean soil counts increased with the sampling depth as shown in Fig. 24.

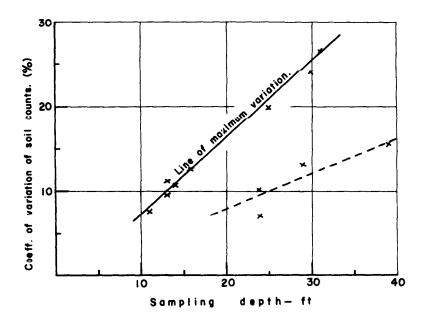


Fig. 24: Graph showing relation between sampling depth and accuracy of counting results of soil samples.