⟨Original⟩

Sorption and Migration Studies of Fission Products for Ground Waste Disposal

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Abstract

The problems of solid waste disposal into the ground in connection with environmental aspects in the vicinity of a site would be very significant, though ground disposal for solid waste is safe and economical method. Studies of the waste-movement and migration of radionuclides (Sr-90 and Cs-137) for the disposal into the ground were performed under laboratory and field conditions.

Affinity of the soils for radionuclide solution was higher than that in the acid solution. The sorption of radionuclides by the soils showed a time-dependent reation. The migration rates of radiostrontium and radiocesium were a range of 3.73×10^{-3} to 10.9×10^{-3} cm/day. The nuclides in the soil migrate much more slowly than the water, probably due to its high exchange capacity.

The observed distribution of tritium was compared with that calculated by a mathematical model based on diffusivity. This study suggests that the tritiated water can be used to trace the movement of ground water.

요   약

부지주변 환경과 관련한 방사성 고체 폐기물의 지중처분 문제는, 비록 안전하고 경제적이라 할지라도, 대단히 중요하다. 지중처분을 위한 방사성 핵종(Sr—90, Cs —137)의 이동과 물의 이동에 관한 연구가 실험실과 야외에서 수행되었다.

방사성 핵종에 대한 토양의 친화력은 알칼리성 용액에서 더 강하고, 흡착은 시간 에 대한 함수관계가 있음을 나타내었다. 방사성스트론티움과 세시움의 이동율은 3.73×10^{-3}—10.9×10^{-3}cm/day 로서, 물의 이동 속도 보다 훨씬 낮은 값을 나타내 었으며, 이것은 아마도 높은 이온이온교환능에 기인되는 것이다.

확산에 기초를 둔 수학적인 도형에 의거 얻은 값과 측정 트리티움의 분포가 비교 되었다. 지하에서의 물의 이동 현상을 규명하는데 삼중수를 추적자로 이용할 수 있 음을 암시하고 있다.

1. Introduction

At the present time nearly three quarters of the worlds' effective energy input comes from oil and gas. But the world nuclear power growth is expected to rise between 227 and 247 GWe in 1981, between 961 and 1,113 GWe in 1991 and between 2,005 and 2,480 GWe in the year 2000[1]. In addition, the development of nuclear

techniques which has inspired a remarkable increase in the industrial, the medical, and the scientific use of radionuclides has been brought in the production of a large volume of radioactive wastes. The ground disposal[2-7] of the various method for radwaste disposal has been in practice and is actively studied in many countries.

Due to the large areas and volumes of soil available in most locations, the potential capacity for sorption of waste is great. And operations by ground disposal have advantages in economy and simplicity. The possibility of contamination of water resources is important in the site selection for the ground disposal of radioactive wastes. Investigations of ground water and the associated migration of radionuclides at the site are necessary to discover any possible hazard from leakage of radioactive materials into the ground.

The migration of radionuclides in the sub-soil is dependent upon two mechanisms: diffusion and dispersion.

The purpose of this work was to make laboratory measurement of the parameters required for the analysis of the migration of radionuclides and to test the applicability of the diffusion theory to explain the observetions of the water-movement.

2. Experiments

1) Sample preparation

The soil samples were taken at three different points (Fig. 1) so that the difference between surface soil (subscript 1) and subsoil (subscript 2) was found. The samples were placed in desicator after these were air-dried and screened through 2 mm sieve (U. S. standard).

2) Physical properties

Particles greater than 2 mm were fractionated by sieve. Air-dried fine soil samples were boiled
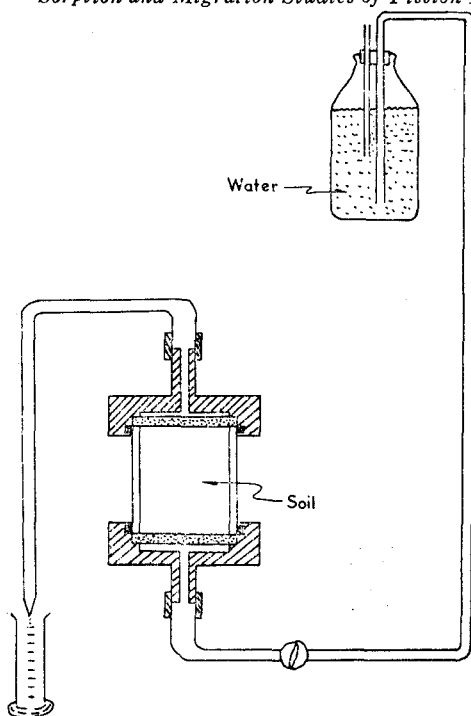

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Fig. 1. K. A. E. R. I. Site

Fig. 2. Shematic diagram of hydraulic condu-
ctivity meauring unit.

with 6% hydrogen peroxide, and soluble salts
were removed by washing with distilled water
and shaked for dispersion. Coarse sand was
fractionated by sieve, and clay and silt portions
were determined by pipet methods[8] using
Stoke's law.

The hydraulic conductivity of soil was meas-
ured by the constant head method[9] using a
device as shown in Fig. 2. Air-dried fine soil
was packed in cylindrical container and saturated
with water which was applied through hydraulic
head at the vertical height of 100 cm at a time
interval measured. The values of hydraulic
conductivity, K, were calculated by Dercy's
equation:

$$K=\frac{Q}{At} \cdot \frac{d}{H}$$

where

Q: amount of water flow

A: cross action of the soil container

t: time of water flow

H: hydraulic head

d: thickness of the soil layer

3) Chemical properties

There were two methods used in analysis of
soils, one of which is decomposed by heating
with HF and $HClO_4$ and the other by fusion
with Na_2CO_3. From the samples prepared by
these, chemical composition (SiO_2, Al_2O_3, and
Fe_2O_3) of the soil was determined by the
method of colerimetry[10-12] with Beckman DU-2
Spectrophotometer.

The total exchangeable metallic cation was
estimated by extraction with NH_4OAc and with
HCl[10].

4) Distribution coefficient

The distibution coefficient Kd does not depend
on the radioactivity of the nuclides but it depends
on the concentrtion of gross ion contained in the
solution and adsorbed on the soil, and pH.

The solution was composed of 0.3M $NaNO_3$,
0.2M NaOH (except the change of pH), and
contained the mass equilvalent of approximately
$1\mu Ci$ of ^{137}Cs per milliliter or $10^{-2}\mu Ci$ of ^{90}Sr
per millillter. One hundred milliters of the
above radioisotope spiked solution with soil
samplesranging from 0.5 to 1.2 gr was employed
to determine distribution coefficient by batch
process.

The change in Kd with increasing contact
time (0.5 hr, 1 hr, 2 hr, ⋯etc.) was calculated
as follows[13]:

$$Kd=\frac{X_1-X_2}{X_2} \frac{ml\ of\ solution}{g\ of\ soil}$$

where

X_1: cpm of the influent solution, and

X_2: cpm of the supernatant solution.

5) Column operation

Ion exchange columns were prepared by pac-
king 35 g of soil sample in a 2.0 cm inner
diameter glass tube to a height of 20 cm. The
soil sample was supported on a glass wool
plug with a wire screen on top. The 35 g
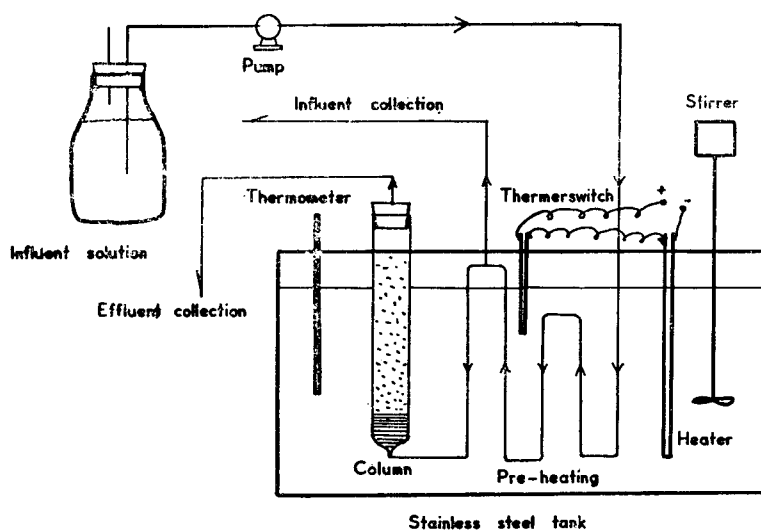portions of soil were soaked in water prior to

Fig. 3. Experimental apparatus for column process

packing in the tubes to allow air to escape from the pores of the soil. The columns were packed under water to exclude air bubbles.

Solutions were pumped downflow through the columns by means of peristaltic action pumps. Effluent samples were collected periodically for analysis. The volume of water above and below the colum was subtracted from effluent volume measurements. The colums were suspended in a controlled temperature water bath maintained at 25°C. The temperature of feed solutions (composed of 0.3 M NaNO3, 0.2 M NaOH, and contained the mass equivalent of approximately 1 μci of ^{137}Cs per milliliter or 10^{-2} μci of ^{90}Sr per milliliter) was controlled at the temperature of 25°C in soils in the water bath before entering the columns (Fig. 3).

6) Water and radionuclides movement

For the estimation of diffusion coeffiicient of water, columns were prepared in a 5 cm inner diameter glass tube to a height of 30 cm. The soil samples (100 g) were compacted on a glass wool plug. On the top of soil a tiny drop of tritiated water was deposited.

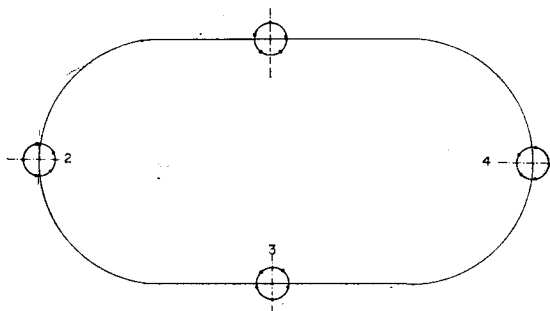After a prechosen time, 1 day and 11 days, the soil plugs at the interval of 1 cm depth

Fig. 4. Tracer injection poiuts and sampling points

were collected. The tritium contained in the water, collected in a multicell vacuum distillation system, was counted in Automatic Liquid Scintillation Spectrometer.

The best way to apply to tracer is with a syringetype injector. 20 point injections at 4 areas, were made along four circle lines (20 cm diameter) at a depth of 0.5 meter (Fig. 4). The amount of trace injection per a time at an individual point was 50 ml of solution contained tracers (^{3}H, ^{90}Sr, and ^{137}Cs) at 2μCi/1. Soil coree were taken with an auger and divided into 10-cm section and collected in wide neck bottles. These bottles are weighed in the laboratory.

Table 1. Mechanical composition of the soils

Soil	Graval 2 mm (%)	G.S 2 mm	F.S 0.2— 0.02 (%)	Silt 0.02— 0.002	Clay 0.002	Tex- ture
A_1	18.8	45.7	25.6	11.9	16.8	SCL
A_2	15.6	28.8	24.4	28.8	17.9	CL
B_1	28.5	59.2	25.0	8.8	7.0	LS
B_2	35.5	59.6	27.9	4.1	8.4	LS
C_1	59.6	69.0	25.5	0.4	5.2	LS
C_2	63.0	63.2	26.2	4.6	6.0	LS

Fine soil

Table 2. Hydraulic conductivities of the soil samples.

Soil	Pore ratio(%)	C (cm/sec)
A_1	50.0	0.111
A_2	48.2	0.079
B_1	53.4	0.265
B_2	53.8	0.283
C_1	55.9	0.576
C_2	54.6	0.394

Table 3. Chemical composition

Soll	SiO_2	Al_2O_3	Fe_2O_3
A_1	21.5	7.6	0.17
B_1	24.5	14.0	0.25
C_1	23.0	2.2	0.17

Table 4. Exthange capacity

Soil	Mep/100g pf soll
A_1	21.5
A_2	25.8
B_1	16.5
B_2	18.2
C_1	13.3
C_2	15.7

The soil samples remaining in the bottles were dried in a multicell vacuum distillation system. The water was collected in small glass bottles and dried soil samples were weighed again.

Table 5. Time dependency of the cesium sorption reaction for soil samples.

Time (hr)	Distribution Coefficient, Kd(ml/g)					
	A_1	A_2	B_1	B_2	C_1	C_2
0.5	65	98	32	36	15	27
1.0	77	116	35	44	15	32
2.0	88	139	41	48	18	38
5.0	110	173	49	57	23	41
10.0	127	208	52	68	27	85
20.0	150	245	61	75	31	63
50.0	184	304	72	91	41	81
100.0	202	384	84	194	48	95
200.0	221	416	93	118	47	102

Table 6. Time dependency of the strontum sorption reaction for soil samples

Time (hr)	Distribution Coefficient, Kd(ml/g)					
	A_1	A_2	B_1	B_2	C_1	C_2
0.5	742	2394	480	463	182	337
1.0	790	2408	485	475	198	341
2.0	838	2443	494	496	204	345
5.0	903	2457	502	505	225	352
10.0	947	2516	518	516	236	359
20.0	991	2560	522	524	252	363
50.0	1006	2615	537	536	274	366
100.0	1068	2624	546	541	292	381
200.0	1143	2709	549	548	293	388

Tritium containig the water was counted in Automatic Liquid Scintillation Spectrometer. 10-gram of the soil was weighed into a 250-ml polyethylene beaker, to which 100ml of distilled water is added and the mixture stirs for 2 hours. The suspended material was then filtered out on a buchner funnel and leached with the distilled water until 200 ml of leachate had been collected. The samples and extracts were then counted by NaI(Tl) Crystal Scintillation Counter and G.M. Counter.

4. Results and discussion

The mechanical composition of the soil analysed is given in Table 1. The samples denoted A, B and C are all sandy and they are dominated by the coarse portion.

It is found that clay portion is increased slightly as the depth is increased.

In general, the hydraulic conductivity of soil is dependent upon the size and distribution of the pore spaces. Table 2 demonstrates the variation of hydraulic conductivities obtained are medium or high[14]. Various theoretical attempts[15] have been made to justify the correlation of hydraulic conductivity with porosity(P). Kozeny[8] has proposed that the value of hydraulic conductivity K varies according to the ratio, $P^3/(1-P)^2$.

Chemical composition of the samples is shown in Table 3. These samples generally contain more amount of silica component than others,
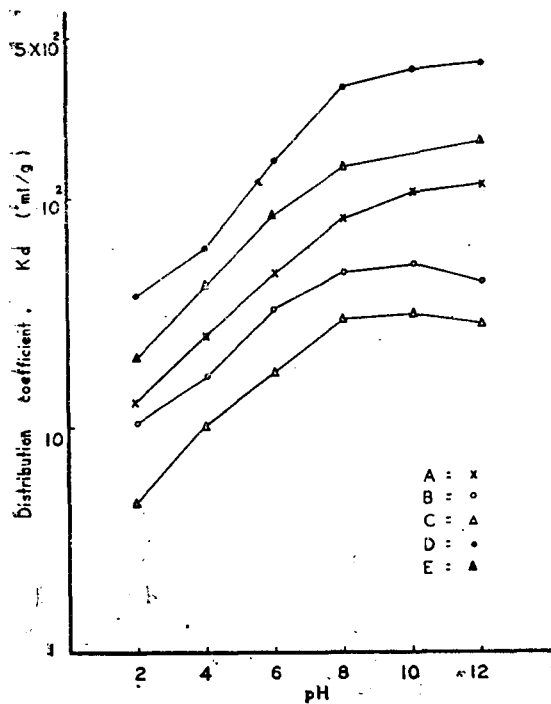
and exchange capacity of A indicates the high distribution in Table 4.

The sorption of strontium or cesium by soils at various initial pH values was evaluated in terms of the distribution coefficient(Kd). Results are given in Fig. 5 and 6 where the Kd was plotted as a function of the initial pH of the soil suspension. Data for some soils for Sr and Cs in dilute solutions increased with the increase in pH. The slight drop in the magnitude of Kd for the B and C soils at pH 10 indicated the loss of sorption sites as the increasing portions of the soils were brought into solution phase with increasing pH.

A second parameter investigated according to slurry techniques was the time dependency of the Sr- or Cs- sorption reaction. The value of distribution coefficient obtained under laboratory condition is summarized in Table 5 and 6. The Sr- or Cs- sorption by the soils showed the
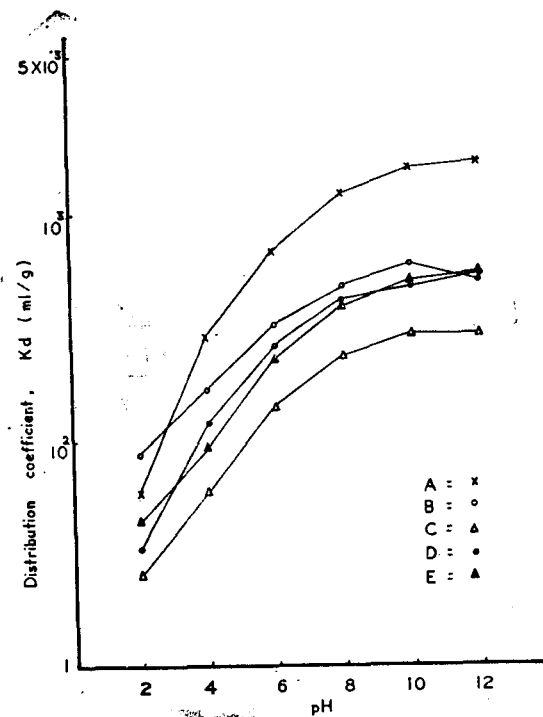

Fig. 5. The sorption of ¹³⁷Cs by soils as a function of pH

Fig. 6. The sorption of ⁹⁰Sr by soils as a function of pH

time-dependent reaction. Kd-value of A-soil was high.

The above rosults indicated that the affinity of soils for Sr or Cs inceased with the increase in clay-fraction, and in the strontium sorption A-soil had the very good affinity.

The results obtained in column operation are shown graphically in Fig. 7-8. Analysis of these data revealed as follows:

(1) Non symmetric break-through curves are at low break-through becoming more diffuse as the percentage of break-through increase. There is an increase in the Sr- or Cs- loading capacity as contact time increases.

(2) The symmetric break-through curves are a result of the more clay-fraction and are suggestive of particle diffusion.

It is assumed that A-site is suitable to dispose into the ground from the above results.

By the above results, A-site was selected for field test. Boring was undertaken with continuous flight augger up to 2 inches in diameter.

As shown in Table 7, underground of the site is composed of, from surface to subsurface, decayed, weathered and fresh rock. The site is consist of two parts; one is excavated part and another is the part of filling-up. In the

Table 7. Depth of each soil or rock layer

unit: meter

Layer Hole Number	Filling Soil	Sandy Clay	Granite	
			Soft	Hard
1		1.6	1.5	1.4
2	2.3	1.1	0.3	1.2
3	2.0	1.1	1.4	5.5
4	1.2		3.8	1.2
Remarks	Filling Soil	Decated Rock	Weathered Rock	Fresh Rock

Table 8. Grain Size distribution and cation exchange capacity with depth

Depth (m)	Grain Size Distribution(%)				C.E.C. meq. per 100 g
	Coarse Sand	Fine Sand	Silt	Clay	
0.5-1.0	39.0	22.9	21.8	16.3	17.5
1.0-1.5	25.8	28.8	29.0	16.4	16.2
1.5-2.0	25.4	30.9	31.8	11.9	11.0
2.0-2.5	23.8	31.9	36.1	8.2	7.9
2.5-3.0	25.6	39.7	27.8	6.9	7.4

former weathered rocks are seen in the surface, but in the latter under the filling-up of decayed rocks, alluvial soil layers about 1.1 m thickness above the granite layers are seen and are composed of sandy clay as weathered rocks.
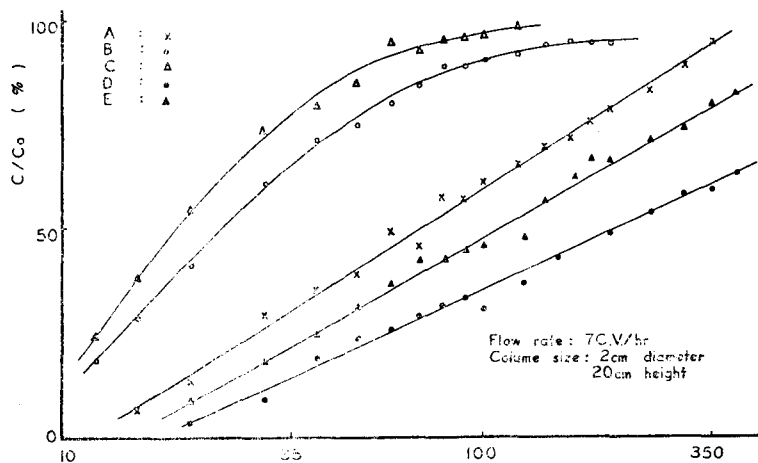
As regards the underground water no under-

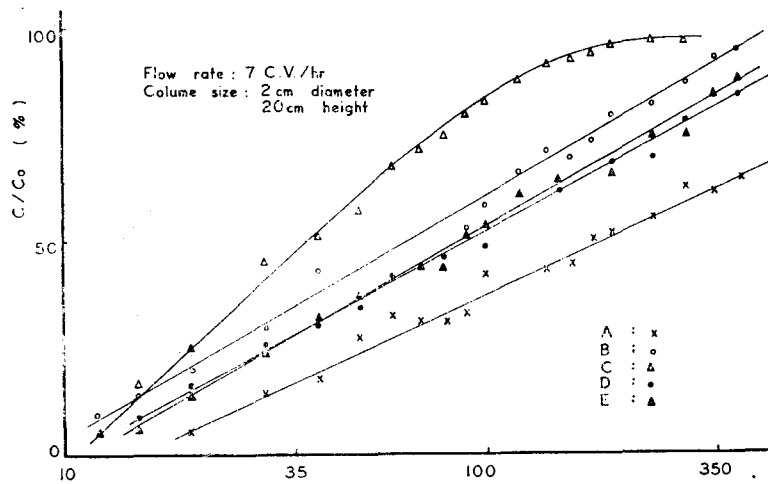Fig. 7. Cesium breakthrough curves for soill sameples with a simulated solution

Fig. 8. Strontium breakthrough curves for soil samples with a simulated solution.

ground water level was found within the depth at hole No. 3 and 4 but at hole No. 1 and 2 underground water was found at 4.2 m and 4.5 m respectively, and the amount of water was a little.

Some of this soil characteristics with depth are listed in Table 8. The soil contains mainly sand and silt but a lower clay content ranging from 7% to 18%. Dation exchange capacity is decreased with the decrease of clay fraction.

Migration of radionuclides in soil is closely related with water-movement in soil. The number of theories of water-movement and migration of radionuclides in soil have been studies by authors[16-19].

Solutions of Fick's law diffusion can be applied to description of the water-movement under a variety of boundary conditions. Convective flow also causes dispersion due to the various paths that the fluid follows as it moves. The distribution of a tracer has been described by formulas relating a diffusivity to time, distance, and physical characteristics of the media. In order to test the applicability of these theories in explaining the observed data, the following equation[20] was utilized:

$$C_x = \frac{Q}{\sqrt{\pi Dt}} \exp\left[-\frac{(x-vt)^2}{4Dt}\right] \qquad (1)$$

where Q is the amount of trace component, C_x is the tracer concentration at time t and at depth x from the injection surface, D is the diffusion coefficient, v is the average intersticial velocity of flow and x is the distance traveled in vertical coordinate.

If the velocity of flow is negligible, the tracer concentration ratio (C_x/C_0) will be given by

$$(C_x/C_0) = \exp\left(-\frac{x^2}{4Dt}\right) \qquad (2)$$

Then, by measuring C_x/C_0, the diffusion coefficient can be calaulated from the equation[2].

The diffusion coefficients of the tritiated water by the column operation were derived and are listed in Table 9. The values were closely aproached to the published values of the diffusion coefficient for water in soils, clays and a matrix of fine glass spheres[21].

When the same amounts of tracers will be injected twice at the time interval, the raio(R_x) of the tracer concentration at depth x to the initial concentration will be derived from the equation[1] as the following:

$$R_x = \frac{\sqrt{t_2}\,e^{-\frac{(x-vt_1)^2}{4_2{}^t{}_1}} + \sqrt{t_1}\,e^{-\frac{(x-vt_2)^2}{4_1{}^t{}_2}}}{\sqrt{t_1} + \sqrt{t_2}} \qquad (3)$$

The existance of the peaks shows that downward water-movement is predominantly by intergranular seepage[22]. From the positions of these
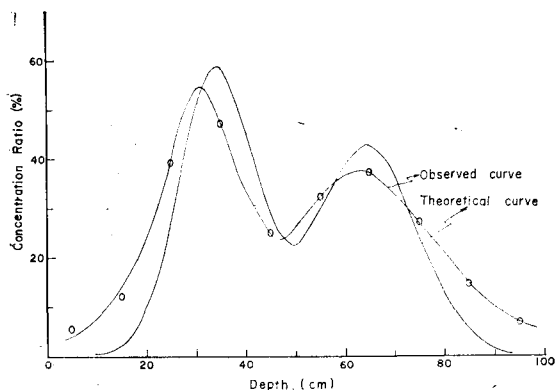
Fig. 9. Distribution Ratio of tritium in the
Point 1

Fig. 10. Distibution Ratio of Tritium in the
Point 3

Table 10. Peakl values of tritium concentration

Samping Point	Travel Time (day)	Ratip(%)	Depth from the Injection Point (cm)	Mean Velocity (cm/day)
1	8	57.0	25	3.57
	15	40.8	50	
2	8	48.2	26	3.43
	15	38.0	50	
3	8	54.2	31	4.28
	15	37.3	61	
4	8	56.1	18	2.43
	15	35.7	35	

Table 9. Diffusion coefficionts of THO at A-site

Run Number	Diffusion Coefficient(cm₂/sec)			
	Point 1	Point 2	Point 3	Point 4
1	2.12×10^{-5}	1.88×10^{-5}	4.18×10^{-5}	1.18×10^{-5}
2	2.43×10^{-5}	2.12×10^{-5}	3.73×10^{-5}	1.36×10^{-5}
3	2.31×10^{-5}	2.21×10^{-5}	4.03×10	1.35×10^{-5}
4	2.25×10^{-5}	1.95×10^{-5}	3.97×10^{-5}	1.16×10^{-5}
5	2.19×10^{-5}	2.05×10^{-5}	4.25×10^{-5}	1.05×10^{-5}
Average Value	2.26×10^{-5}	2.04×10^{-5}	4.03×10	1.21×10^{-5}

peaks, the mean velocities of downward water-movement are 3.57 cm/day at point 1 and 4.28 cm/day at point 3 as shown in Fig. 9 and 10. The first peaks on the observed profiiles are slightly lower than those on the calculated one, presumably due to the adsorption of the tritium on the soil. These show that the equation is one possible interpretation of these profiles.

It is due to the formation of fresh rock layer from about 2 meter depth that peaks were observed at the point 2 and 4. If we assume that the sharp peak is plotted on the injection point, the mean velocity of downward
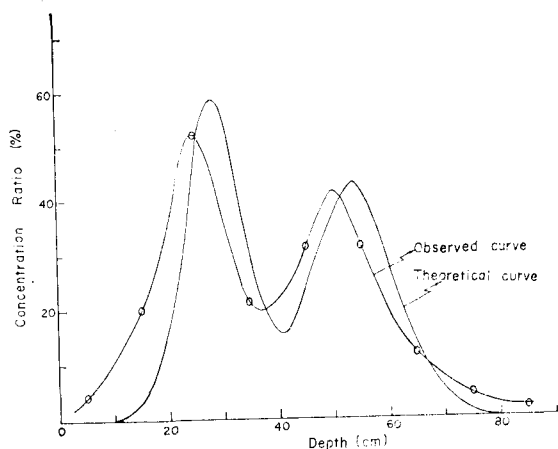
water-movement at the point 2 and 4 3.43 and 2.43 cm/day, respectively (Table 10).

By Marter and Aiken[23], a percolation rate of 21.2—27.3 cm/day has been reported for saturated flow but only 3.0 cm/day or less for unsaturated flow.

Many theories on the migration of radionuclides have been developed for radionuclides migrating unidirectionally through homogeneous geological formations. Inoue[24], however, developed a theoritical equation[4] which could be used for the migration of radionuclides through actural field strata.

$$\frac{V}{V_A}=1+\frac{1-P}{S\cdot P}\rho K_d \tag{4}$$

where V is the movement of ground water, V_A

Thable 11. Migration rates of radiostrontium and radiocesium

Sampling Point	Porosity	Density (g/cm³)	Saturation factor	Distribution Coefficient(ml/g)		Migration Rate (cm/day)	
				Sr-90	Cs-137	Sr-90	Cr-137
1	0.484	2.42	0.82	254	113	4.46×10^{-3}	10.0×10^{-3}
2	0.479	2.90	0.82	216	121	4.12×10^{-3}	7.35×10^{-3}
3	0.431	2.65	0.73	239	82	3.73×10^{-3}	10.9×10^{-3}
4	0.512	2.23	0.86	124	98	7.90×10^{-3}	9.99×10^{-3}

is the migration rate of a radionuclide A through porous media, P is the porosity of the soil, ρ is the density of the soil, K_d is the distribution coefficient of the nuclide A, and S is the saturation factor.

Radionuclides in the solution extracted with distilled water were not detected and then migration rates of the nuclides were measured by the field test. It is estimated to be due to the lower activity and the short travel time of tracers (Sr-90 and Cs-137). It is also considered that all of these nuclides were fixed on the soil surfaces. After 300 days about 95% of the total activity applied is retained in the first 10 cm of the soil. Between a depth of 10 and 20 cm a slight concentration of radionuclides is noticeable.

Frederikson et al[25] showed that 12 months after contamination with ^{106}Ru, ^{137}Cs and ^{144}Ce at least 80% of the contamination was found in a layer 2.5 cm below the surface. Peirson and Salmon[26] pointed out that 80% of the total world-wide fall-out can be found in a layer of 7 cm below the surface.

The migration of radionuclides was determined by equation[4] and the results are listed in Table 11. These approachs to the previous data[27]. The nuclides at each point migrate much more slowly than the water. The distribution coefficients of the nuclides showed the same trend as shown in Table 5 and 6. The results indicated that the affinity of the soils for strontium is higher than that for cesium. It is also shown

that strontium at each point migrate more slowly than cesium. The slight downward movement of the nuclides in this soil is probably due to its high exchange capacity.

6. Conclusoin

This study showd that the tritiated water can be used to trace the movement of ground water. The observed distribution of tritium compared with that calculated by a matheatmical model based on diffusivity process.

The migration rate of radionuclides (Sr-90 and Cs-137) in the ground is very slow, about 95% of the total activity can be found in the first 10 cm below the injection surface.

These facts shown clearly that the migration of nuclides is effectively restricted by the exchange capacity of soil to hold radionuclides although the soil has less exchange capacity in unit volume than that of an artificial ion-exchange resin.

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