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TRITIUM CONTAINMENT AFTER BURIAL OF CONTAMINATED SOLID WASTE

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0565 Reference

ABSTRACT

Asphalt has been used since 1970 as a means of containing tritium in solid waste disposed of by shallow earth burial in volcanic tuff. As applied, the asphalt has been ineffective in limiting or preventing tritium migration from the waste burial site. Similarities observed between tritium distribution patterns and orientations of joints in the tuff indicate that the migration occurs primarily as movement of tritiated water vapor through joint systems. Modified asphalt-containment techniques now being implemented are described.

INTRODUCTION

The Los Alamos Scientific Laboratory (LASL) has used on-site shallow earth burial as the means of disposal of solid radioactive wastes generated in various research and development programs since the beginning of the Laboratory in the mid-1940's. The Los Alamos Laboratory, situated in a semi-arid region of the mountains of northern New Mexico, is considered to have very favorable climatologic and geologic conditions for disposal of radioactive materials in this manner. Radionuclides disposed of in solid wastes include significant amounts of transuranics (mostly plutonium and americium), uranium, fission products, activation products, and tritium. All investigations of the radioactive solid-waste disposal areas have revealed that, with the exception of tritium, no migration of these radionuclides has been detectable.

Tritium has a relatively short half-life of 12.3 yr, compared to half-lives of tens to thousands of years for other radionuclides. In addition, the recommended concentration guides for exposure to tritium in both air and water are considerably higher than for other radionuclides of environmental concern.[1] Nevertheless, because of the high mobility of tritium in biological systems,[2] there is a legitimate need to define and minimize the quantities of this radionuclide entering the environment from disposal areas, or from any other sources.

Migration of tritium from its burial ground location was detected at the presently active disposal site in 1970. Research being carried out at that time in Los Alamos identified asphalt as a material capable of slowing or limiting migration from waste. Consequently, attempts have been made since then to contain with asphalt most of the tritium being disposed.

In this paper, the actual effectiveness of the asphalting techniques used to contain tritium in solid wastes disposed of by burial is discussed, and mechanisms by which tritium has migrated from burial sites in the Los Alamos environment are identified.

PHYSICAL SETTING

The Los Alamos Scientific Laboratory is located in north central New Mexico, along the western margin of the Rio Grande Basin at an elevation of about 2200 m. The Laboratory and nearby townsite are situated on the Pajarito Plateau, a topographic high, ~600 m above the Rio Grande. The plateau is composed of moderate-to-densely welded rhyolitic tuffs of the Bandelier Formation.[3] The tuff is broken into blocks by numerous joints at intervals of 1 to 2 m. The tuff deposits overlie sediments of the Rio

9191

Grande basin. The regional water table is located in these sediments, at depths of 200 to 300 m below the surface of the plateau. The plateau is cut by numerous east-west canyons that open into the present Rio Grande drainage at intervals of one to several kilometers. The Laboratory facilities are primarily located on the tops of the resulting fingerlike mesas. Perennial streams flow only in the upper end of many of the canyons.

Los Alamos has an average annual precipitation varying from 300 to 500 mm on the Pajarito Plateau. Potential evapotranspiration is ~600 mm in the area.[4]

The residual soils on the Bandelier tuff are quite thin, typically <1 m thick, and have a thin clay horizon at the soil-rock contact which restricts vertical movement of water. The tuff has a relatively low hydraulic conductivity when saturated with moisture and the conductivity decreases rapidly with reduced moisture content.[5] The combined effect of the precipitation/evaporation ratio and the relative impermeability of the soil and rock material significantly restricts the quantities of moisture penetrating below a few meters in the tuff. This is substantiated by numerous data on distributions of moisture with depth in the tuff, obtained by both neutron logging techniques and from holes augered into the tuff.[5-7] These data indicate that the moisture content of the tuff is generally 5% or less, by volume, at depths >4 m, even in areas where the soil cover does not exist or has been removed by excavations.

TRITIUM WASTE DISPOSAL AT LASL

Shallow earth burial of radioactive solid wastes at LASL has involved the use of pits and, more recently, drilled shafts. Typically, the pits are 8 m or less in depth. The disposal shafts vary from about 8 to >20 m in depth and from 0.6 to 1.8 m in diameter. All of the radioactive solid-waste disposal sites used by the Laboratory have been located on the surface of mesas, on the Bandelier tuff. The site presently being used, referred to as Area G, is located on Mesita del Buey, ~8 km from the central laboratory area.

The very low moisture content of the tuff results in little, if any, moisture movement below a depth of a few meters, with the bulk of that occurring in the vapor phase. As a result, dissolution and subsequent migration of the radionuclides in the disposal area is minimal to nonexistent. This has been verified continually by analysis of innumerable samples collected, as standard procedure, during the excavation of additional shafts and pits in the disposal areas. Tritium is a notable exception to this. Occurring principally as tritiated water, this radionuclide is nearly as free to move as the water itself. Small quantities of tritium gas, which may be disposed of by burial in waste, convert very quickly to HTO on exposure to the soil moisture.[2]

Tritium-contaminated solid wastes at LASL have primarily consisted of tritiated water, oils, laboratory apparatus, tritium contained in vacuum-system components and traps, cylinders, shipping canisters, and bound tritium as in uranium tritide. Existing radioactive solid-waste disposal records indicate that of a total of ~220,000 Ci disposed of by the Laboratory as of 1975, 162,000 Ci (73.6%) has been tritium.

Prior to mid-1958, all tritium-containing wastes were disposed of by burial with other wastes in the disposal pits. Since that time, and until November 1973, unlined 0.6- to 1.8-m-diam × 8- to 20-m-deep shafts were used. Packaging of this waste until late 1970 ranged from plastic bags and cardboard boxes to a variety of sealed metal containers such as steel drums and small paint-type cans.

In 1970, the Industrial Liquid Waste Group (Group H-7) at LASL carried out an investigation of tritium loss from cement paste blocks.[8] Their primary objective was to develop an acceptable means to dispose of water containing millicurie quantities of tritium per liter. The tritium-containing water was combined with Portland cement to form a paste which was molded into blocks which were then coated with the various test materials. Leaching of tritium from the coated blocks was measured. In these studies, an asphalt coating of 1.9-cm thickness provided the most effective barrier to tritium migration out of the block. Less than 0.01% of the contained tritium escaped over a 56-day immersion of the block in water. A block dipped in hot asphalt, which provided a much thinner coating, lost only 0.8% of the contained tritium over the same period.

Coincidental with this laboratory investigation, Purtymun[9] detected migration of tritium into the tuff surrounding disposal shafts at the active waste disposal site. As a direct result of these two investigations, in November 1970 the Los Alamos Laboratory began packaging tritium wastes for disposal into 210-liter mild steel drums with asphalt-coated interiors. In many instances such wastes were prepackaged in a 115-liter steel drum which was then placed into the coated drum. In most instances, however, the only seal for the drum lid was the rubber gasket provided with the drum. More recently, some tritium wastes have been prepackaged in the 115-liter drum which has then been completely encased in asphalt in the 210-liter drum.

Since November 1973, in addition to these packaging procedures, the walls and bottom of the disposal shafts for tritium waste have been coated with a 1- to 2-cm coating of asphalt. The primary motivation for this was an attempt to seal the many fractures in the tuff walls of the shaft, as it was believed that such fractures provided possible pathways for tritium migration.

The location of disposal shafts used for disposal of tritium-contaminated waste is indicated in Fig. 1. The previous study of tritium migration indicated that shafts 11, 12, or 13 were significant sources of tritium. These shafts, together with shafts 1-10, received waste without asphalt containment. Shafts 50 and 59 have received tritiated waste with additional asphalt containment.

A summary of the tritium disposals into these shafts is presented in Table I. For several disposals of tritium wastes the disposal records are incomplete, as values for the tritium content were not recorded. Through knowledge of the waste and its origin, estimates of the tritium contents have been made. These are listed in the column labeled "Additional Estimated Ci." The last column in Table I gives an estimate of the maximum quantity of tritium that might reasonably be expected to be available for migration away from these shafts. These values were derived by subtracting from the total tritium estimated as present, that tritium contained in cylinders, shipping canisters, and other metal containers that are reasonably expected to remain intact and leak-free following burial.

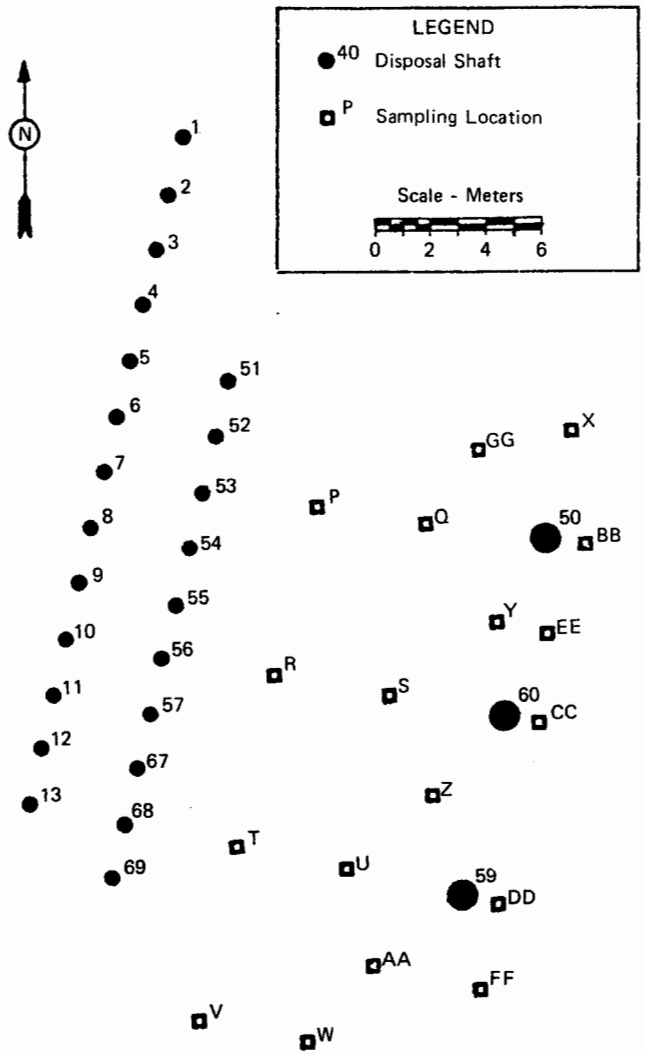

Fig. 1. Location of disposal shafts and sampling holes, Area G.

FIELD INVESTIGATIONS

Shafts 50 and 59 are substantially separated from older shafts used for tritium disposals, and

TABLE I

Summary of Tritium Disposals at Study Site

Shaft No. (use dates)	Recorded Disposals (Ci)	Additional Estimated (Ci)	Total ^3H (Ci)	Potential ^3H Available for Migration (Ci)	
1-10 (8/66-8/69)	6000	800	6800	700	No Asphalt Containment
11-13 (7/66-4/70)	70	300	370	350	
59 (11/73-5/74)	2600	600	3200	1800	Asphalt Containment
50 (6/74-present)	7800	100	7900	7700	

both have received only wastes to which the asphalt-containment techniques were applied. Their location provides an opportunity to determine the effectiveness of the new disposal techniques.

The volume concentration of tritium in the tuff is greater near a source of tritium than at some distance from it. Thus, the tritium distributions in the tuff can be used to identify the various shafts from which tritium migration is occurring. The movement of tritium within the tuff may result from self-diffusion along tritium concentration gradients or through convection by air or water movements within the tuff. The presence of open joints will have an effect on all three mechanisms. It was thus necessary to evaluate the extent and orientation of joints within the tuff at the disposal site.

The distribution of HTO in the tuff within the disposal field was determined by drilling holes with a 4-in. power auger on a predetermined grid. The location of sampling points for this study is indicated in Fig. 1. Composite samples of tuff cuttings were collected for each 1.5-m depth interval, to depths of 12 m, and double bagged in plastic. Moisture was collected from the samples using a heat-distillation technique and the moisture samples were assayed for tritium using a Beckmann liquid-scintillation counter. There are two principal sources of error in the data, sample variation and counting errors. The latter are on the order of ±3%. The extent of sample variability was determined through collection of replicate samples and comparison of the measured HTO content. Errors ranged from ±1 to ±40%, with a mean of ±16%. Tritium and moisture concentrations at various depths, as determined by this study, are presented in Table II.

The principal source of data on orientation of the joints is the measurements made of the strike and dip in the exposed faces of excavations in the tuff. Observations were made, as part of this study, of the orientation of joints intersecting shafts 50-57 and 67-69. The joints are generally vertical but occasionally curve to dips of 60 deg or less. Near the surface the joints are filled with soil material or with weathering products derived from the walls of the joints.[10] At depths >5 m, the joints are generally open and may reach widths of several centimeters. Few data are available on the average length of the joints as they are difficult to trace on the surface. Exposures of joint faces on the walls of excavations have extended up to 40 m. Joint frequency varies considerably but, on the average, joints are spaced $1\frac{1}{2}$ to 2 m apart. A summary of the orientations of joints observed on the walls of burial pits at Area G is presented in Fig. 2 (Refs. 11 and 12). Orientations of joints observed in the walls of disposal shafts lie within this distribution, with two principal directions dominant; N 70 deg W, and N 50 deg E.

INTERPRETATION AND DISCUSSION

Tritium and moisture distributions surrounding disposal shafts 50, 59, and 60 for two depth intervals are presented in Figs. 3 and 4. For comparison, tritium distributions surrounding shafts 11, 12, and 13, as determined by Purtymun,[9] are presented in Fig. 5.

Shafts 50 and 59 have received tritium-contaminated waste since 1973, as indicated in Table I. Both shafts were asphalt lined, and asphalt containment was applied to the packaging of wastes placed in the shafts. Shaft 60 has received no tritiated waste. The influence of shafts 50 and 59 on the tritium distributions is apparent, with elongations of the isopleths in directions coincident with major joint orientations indicated in Fig. 2.

Comparison of Figs. 3, 4, and 5 indicates that the concentration of tritium in the tuff surrounding shaft 59 was considerably higher at the time of the present study than were the concentrations surrounding shafts 11-13, as determined by Purtymun.[9] Approximately the same time period elapsed in both instances between the placement of the waste and the collection of samples. The total tritium present in a given depth interval can be estimated from Figs. 3, 4, and 5 by determining the area within each isopleth interval, and multiplying by the mean concentration for that interval. A summary of those calculations for the 6- to 7.5-m depth interval is presented in Table III.

Similar comparisons can be made for the other depth intervals surveyed in this and the previous study. These estimates indicate that approximately ten times as much tritium has migrated out of shaft 59 as out of shafts 11-13. As indicated

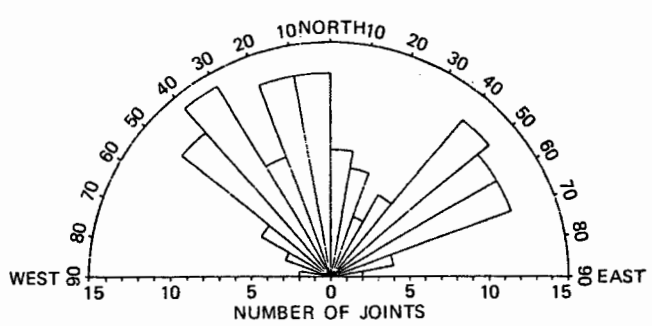

Fig. 2. Orientation of joints in Bandelier tuff adjacent to disposal shaft field, Area G.

TABLE II

Tritium and Moisture Content of Tuff Surrounding Shafts 50, 59, and 60

Hole	Depth (m)							
	0-1.5	1.5-3	3-4.5	4.5-6	6-7.5	7.5-9	9-10.5	10.5-12
P Mois[a]	8.9	8.1	2.2	1.0	<1.0	<1.0	<1.0	<1.0
P ^3H[b]	810	9 200	6 600	6 200	<10 000	<8 000	<5 000	<5 400
Q Mois	7.9	8.1	2.2	1.8	1.0	1.0	1.0	1.0
Q ^3H	1 400	24 000	16 000	46 000	20 000	20 000	33 000	350 000
R Mois	7.7	7.2	4.7	3.6	1.4	<1	<1	<1
R ^3H	310	1 700	3 600	2 800	2 200	<2 000	<2 000	<2 000
S Mois	10.2	8.9	6.6	2.9	3.0	1.8	1.2	---
S ^3H	490	1 000	2 300	1 200	2 700	2 700	5 800	---
T Mois	9.4	15.7	5.5	4.0	2.8	1.0	0.3	<1
T ^3H	240	2 200	990	1 200	1 600	680	160	<100
U Mois	11.4	10.2	6.6	6.0	5.7	5.8	7.3	5.7
U ^3H	9 400	91 000	59 000	44 000	34 000	16 000	39 000	22 000
V Mois	9.9	7.8	6.0	4.6	4.1	2.8	2.2	1.6
V ^3H	400	3 000	4 700	8 200	14 000	8 400	4 800	3 800
W Mois	18.9	6.5	5.5	4.7	3.2	2.2	1.8	2.5
W ^3H	1 200	5 100	7 800	10 000	5 900	3 400	2 100	2 200
X Mois	11.0	10.1	7.2	6.3	6.3	5.4	3.4	2.1
X ^3H	2 400	8 900	56 000	52 000	58 000	63 000	88 000	80 000
Y Mois	4.3	5.8	5.1	4.0	3.0	---	---	---
Y ^3H	820	69 000	47 000	60 000	50 000	---	---	---
Z Mois	11.6	9.1	6.4	---	6.0	---	1.0	1.0
Z ^3H	1 800	7 100	8 100	---	28 000	---	4 000	3 500
AA Mois	12.0	11.5	10.7	6.1	5.3	3.4	<1	<1
AA ^3H	24 000	150 000	360 000	21 000	25 000	8 500	<8 000	<8 000
BB Mois	12.8	11.7	7.6	6.3	6.4	5.1	2.3	1.3
BB ^3H	1 800	23 000	120 000	100 000	40 000	55 000	55 000	34 000
CC Mois	9.8	11.3	7.1	5.8	4.0	3.3	5.5	9.5
CC ^3H	720	1 900	5 600	1 600	2 500	3 600	120 000	14 000
DD Mois	13.2	10.0	6.7	7.1	6.2	5.4	3.0	1.7
DD ^3H	6 300	52 000	37 000	24 000	12 000	3 800	1 300	2 100
EE Mois	12.0	11.1	10.4	10.4	6.6	6.2	5.7	6.2
EE ^3H	1 900	2 800	5 800	6 200	16 000	60 000	120 000	93 000
FF Mois	15.2	15.1	11.2	9.9	7.0	6.3	6.8	6.5
FF ^3H	4 800	18 000	180 000	170 000	180 000	78 000	22 000	23 000
GG Mois	9.6	9.5	10.5	10.3	6.4	6.5	6.3	6.4
GG ^3H	1 900	1 900	13 000	13 000	32 000	39 000	39 000	34 000

[a]Moisture content, percent by weight.
[b]Tritium concentration, pCi/cm^3 tuff. Assumes uniform bulk density of 2 g/cm^3. Regional background values are 2 to 10 pCi/cm^3.

in Table I, the tritium presumed to be available for migration in shaft 59, if the asphalt containment had not been applied, is more than five times that estimated to be available for migration from shafts 11-13. Thus, on a percentage basis, significantly more tritium has migrated out of shafts where asphalt containment was used than from shafts where no asphalt was applied. Undoubtedly many factors contribute to this difference, including differences in waste form and packaging prior

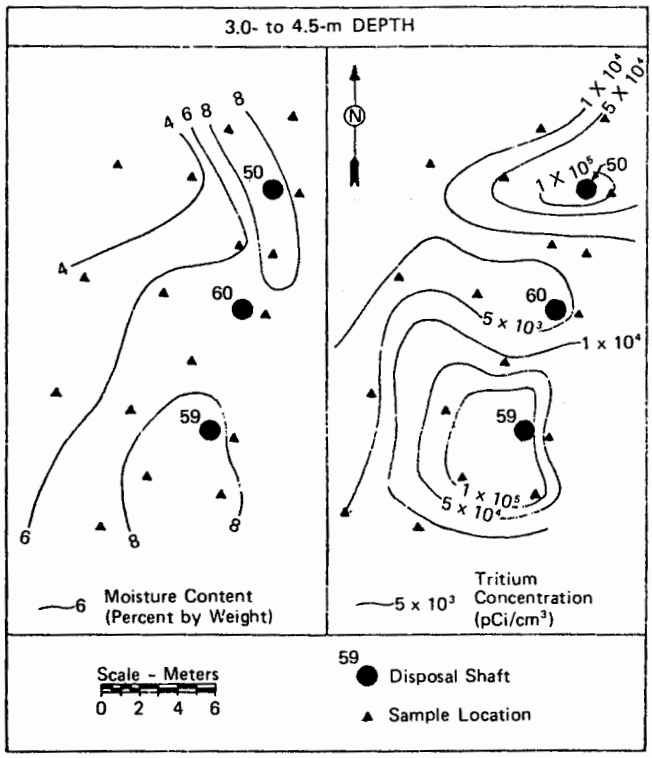

Fig. 3. Moisture and tritium distributions in 3.0- to 4.5-m depth interval, surrounding shafts 50, 59, 60 (from data presented in Table II).

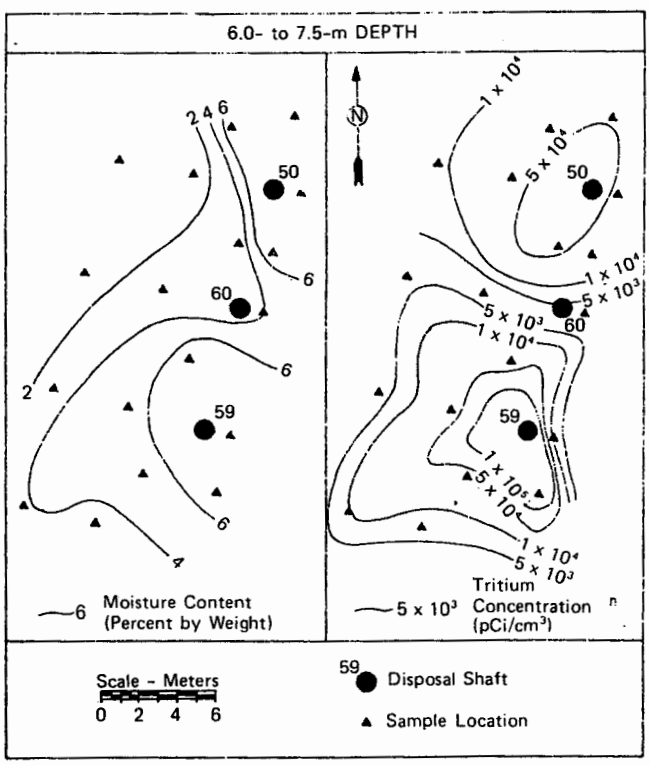

Fig. 4. Moisture and tritium distributions in 6.0- to 7.5-m depth interval, surrounding shafts 50, 59, and 60 (from data presented in Table II).

to asphalting, variabilities in permeability of the tuff, and differences in the size and number of fractures intersected by the various shafts. However, the asphalting was intended to counteract and override these variabilities. It is concluded that the asphalting techniques, as applied in the past, did not significantly reduce the migration of tritium away from the disposal shafts.

The shafts also appear to serve as a moisture source to the tuff, particularly at depth. This is probably because of the entrance of precipitation

TABLE III

Tritium in 6- to 7.5-m Depth Interval Surrounding Disposal Shafts

Shaft	Minimum Isopleth Used (pCi/cm^3)	Total Area Inside Isopleth (m^2)	Tritium Within Isopleth (Ci)
11-13	1000	115	1.2
59	5000	150	11

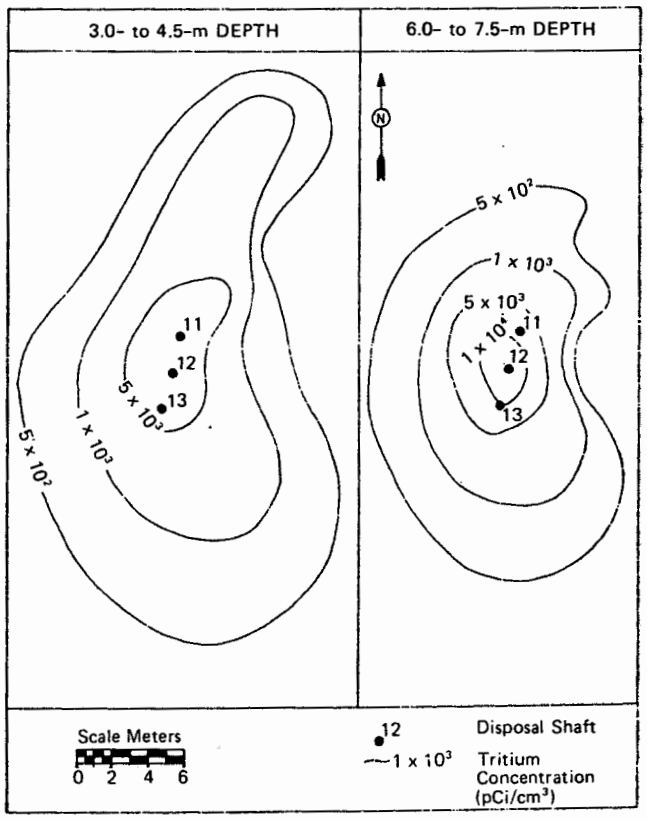

Fig. 5. Tritium distributions for two depth intervals, in tuff adjacent to shafts 11-13 (after Purtymun[9]).

into the shafts, principally by flow down the sidewalls. The shafts are covered between waste deliveries but are not totally sealed against the entrance of surface runoff. Elongation of the tritium isopleths and, to a limited extent, of the moisture isopleths in the direction of major joint orientations indicates that the joints are providing preferential pathways for both tritium and moisture migration. Under unsaturated conditions, open joints provide a barrier to the movement of water in the liquid state but not for water in the vapor state. Thus, tritium migration would appear, as anticipated, to result from the movement of tritiated water vapor through the tuff.

SUMMARY AND CONCLUSIONS

Migration of tritium from subsurface disposal shafts at LASL occurs through movement of tritiated water vapor through rock material. The use of asphalting techniques, as they have been applied to date, has not significantly reduced the migration of tritium away from the disposal shafts. This is more likely because of inadequacy of the encapsulation techniques than to the ineffectiveness of asphalt as a containment medium. The principal pathway for migration out of the disposal shafts is the joints that intersect them. Asphalt coating the interior of the shafts appears to be ineffective in sealing these joints. Consequently it is anticipated that this procedure will be discontinued.

Currently, two modifications of the methods of tritium waste packaging are being utilized at Los Alamos in an attempt to provide better primary containment of the tritium. One technique involves the use of an asphalt-type roofing compound as a lid sealant on both the inner 115-liter drum containing the waste and on the 210-liter asphalt-coated drum into which the smaller drum is placed. In the other, a 115-liter drum is suspended inside a 210-liter drum, and the void space between the two is filled with asphalt. Following the filling of the 115-liter drum with waste and sealing of the inner drum, additional asphalt is used to complete encasement of the waste. These modified packaging techniques will be evaluated separately, either in the laboratory or in a burial location, by monitoring for tritium releases from drums containing tritiated waste.

ACKNOWLEDGMENT

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